Technical Session III Implementation Issues

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Processed Engineered Fuels Derived From Paper and Plastics -- Techno-Economic Factors and Regulatory Issues in a Competitive Market

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INTRODUCTION

Plastics have gained a prominent position in modern society because their unique properties, cost-performance and versatility. They contribute to almost every aspect of modern life including transportation, recreation, medicine, packaging, appliances, furniture, textiles and building and construction. The diverse properties of plastics allow them to play a major role in providing solutions for technical progress as well as solutions for everyday life.

Plastics also make a significant contribution to effective resource management and conservation, reducing our use of natural resources and helping to minimize waste. By choosing plastics, manufacturers, designers, distributors and retailers in every sector have been able to respond to the challenge of preventing excess use of resources and, at the same time, bring increasingly sophisticated products to the market. Resource conservation seeks to maximize efficiency and minimize waste from a product's manufacture, use and disposal. It involves more than managing and minimizing waste, however; it also includes taking advantage of effective life-cycle resource management at local and global levels. There are many ways that plastics contribute to effective resource management. From manufacture to use, to recovery, to waste management, plastics help conserve resources. Not only are plastics energy efficient, but they often create less waste than other materials. They can be recovered, recycled and safely landfilled.

Plastics play an important role as a potential source of energy. Since they are manufactured from oil and natural gas, plastics can be viewed as "diverted" energy that is reclaimed in an energy recovery process. Recovering energy from post-consumer plastics is important for two reasons. First, plastics contain the highest energy value of any component of the municipal solid waste stream. Second, practical, technical and economic barriers limit the sensible reuse and recycling options of any material. By committing high-energy materials like plastics to land disposal, a significant energy resource is lost.

One objective of the American Plastics Council (APC) is to explore and extend the benefits of plastics that are not currently being realized and utilized. An important goal is to help ensure that all economic and environmentally responsible recovery options are available for plastics. By providing information about the environmental and economic impacts, positive and negative, of each municipal solid waste management and resource recovery option, we can make more informed decisions and achieve the goal of effective resource conservation.

This paper reviews the energy recovery option for post-use plastics. It discusses plastics' role in modern waste-to-energy (WTE) systems and then reviews some recent work exploring the use of plastics as alternative fuels. The emphasis is on solid fuels, often referred to as process engineered fuel or PEF. The liquefaction and gasification of plastics offers additional opportunities for resource recovery, but this subject is only briefly discussed in this paper.

OVERVIEW

Figure 1 summarizes three basic options for the recovery of energy from post-use plastics. They are municipal solid waste combustion or waste-to-energy; process engineered fuel (PEF) derived from plastics or combinations of plastics and paper; and liquid or gaseous fuels derived from the liquefaction or gasification of post-use plastics. The end product from all three processes is useful energy. The first option can be categorized as commercial and mature, the second as commercial and embryonic, especially with regard to the use of plastics; and the third as developmental. On the basis of these three options and the expanding need for energy worldwide, energy recovery from post-use plastics is expected to grow as a significant component of integrated resource management.

WASTE-TO-ENERGY (MUNICIPAL WASTE COMBUSTION)

According to the most recent U.S. EPA report on municipal solid waste titled *Characterization of Municipal Solid Waste in The United States: 1995 Update*, ¹ plastics make up 9.5 percent by weight (19.8 million tons) of municipal solid waste. On this basis, plastics contribute almost 30% to the fuel

value of MSW. In combination with the non-recycled paper content of MSW, the combined contribution is more than 50% of the fuel value (Figure 2).

The same EPA study found that in the United States, 20% of MSW, after recycling and composting, is combusted--essentially all with energy recovery. The 20% figure is not often cited. The more common convention in the United States is to report the percentage for a given disposal or recovery technology applied to the total of all MSW generated. On this basis, the percent of MSW combusted is about 15.5% according to both EPA and Integrated Waste Services Association (IWSA) data.² It is often unclear on which basis European and Japanese data are being presented. Nevertheless, many countries such as Denmark, Switzerland, and Japan, and some regions of the United States, presently recover the energy from over 50% of generated MSW. Table 1 summarizes information from the Integrated Waste Services Association (Washington, DC) on municipal waste combustion, which confirms that more than 97% of MSW combustion in the United States involves energy recovery.

A recent modeling study suggests that there is significant potential for further growth of the waste-to-energy industry in metropolitan areas across the United States under the assumption that the minimum viable resource recovery plant throughput is 400 tons per day of MSW. This was essentially a hypothetical landfill diversion study. The study indicated that up to 76% of the U.S. population could effectively be served by modern mass burn technology compared to about 16% today. With 76% of the population served, approximately 58% of generated MSW would be managed in waste-to-energy facilities. In practice, of course, marketplace economics are often the controlling factor as to which waste management option is used.

The same study examined certain economic assumptions which could impact the viability of WTE in the future. Figure 3 shows a theoretical relationship between the calculated tipping fee which would need to be charged by a newly constructed WTE plant to break-even as a function of electricity sales price (effectively revenue). For comparison, Figure 3 also shows the range of average regional landfill tipping fees across the U.S. According to the assumptions of this model, relatively high electricity sales revenues are needed for WTE costs to be competitive with average landfill tipping fees. It is important to note, however, that this model is for new plants and assumes that all of the ash would be landfilled as an expense, rather than beneficially used.

As noted above, plastics provide an important fuel source for the waste-to-energy process. Indeed, in the U.S. today, roughly one out of every five pounds of post-use plastics are used for energy recovery in modern waste-to-energy plants. A recent study in Würzburg, Germany involving a mass burn municipal waste combustor equipped with baghouse and carbon injection demonstrated that mixed plastics in the MSW stream improve burn-out of gases and solid residue without adversely affecting ash or emissions. ⁴ As plastic loadings were increased during the tests, CO and SO₂ emissions were reduced. In addition, higher polymer content, including specific addition of polyvinyl chloride, did not result in an increase in dioxins and furans. This finding supports the general conclusion of a recent ASME study that dioxin emissions from modern waste-to-energy plants do not correlate statistically with the quantity of chlorine in the feed. ⁵ In fact, both the findings of the Würzburg study and the International Ash Working Group Report ⁶ indicate that modern waste-to-energy plants are a net destroyer of dioxins.

Today, waste-to-energy technology is playing an important role in utilizing the energy content of plastics and other polymeric materials, and there are additional opportunities for the future. Recently, APC completed a study to assess the energy recovery option for automotive shredder residue, the byproduct of the automobile shredding operation. ⁷ The energy content of ASR ranges from about 4000 to 11,000 Btu/lb depending on the degree of processing. The study looked at mass burn technology, RDF, industrial boilers, cement kilns, and emerging gasification technologies, and concluded that cofiring ASR with MSW in a modern mass burn waste-to-energy plant offered the most practical means to capture the inherent energy of this material.

PROCESS ENGINEERED FUEL

Basic Definition

Process engineered fuel (PEF) can be defined as a processed solid fuel, made from segregated plastic and paper, derived from industrial, commercial, and residential sources, for use by utilities and industry. ⁸ The term paper is used broadly to refer to a wide variety of non-recyclable paper, boxboard, corrugated, and other cellulosic-based feedstocks. Plastic feedstocks can be in rigid, foam, or film form. Figure 4 shows the sourcing options, fuel processing step, and markets for PEF. PEF can be produced and marketed in both densified and fluff forms.

The term Process Engineered Fuel has not yet gained wide acceptance in Europe. The terms Packaging Derived Fuel (PDF) and Plastics Packaging Derived Fuel (PPDF) have been proposed ⁹ but not broadly accepted. "Alternative fuel" is considered an appropriate generic term. In practice, PEF feedstocks often go beyond packaging-derived materials. When derived from post-use materials or from industrial, agricultural, or forest based products, or related industrial process streams, e.g., certain biofuels, these fuels all fall under the broad category of renewable fuels. The production and use of PEF is often referred to as fuel recovery rather than energy recovery.

Process engineered fuel should not be confused with Refuse Derived Fuel (RDF). A recent paper proposed a classification system to differentiate PEF, RDF, and MSW based on certain fuel properties. ¹⁰ A basic distinction can be made on the basis of higher heating value with a lower limit of 6500 Btu/lb suggested for PEF (Table 2). This comparison is useful, but the definition of PEF provided above suggests that only in certain cases would PEF feedstocks be related to MSW or RDF. PEF should be viewed as a manufactured fuel not a waste. Definitions and terminology related to PEF are further discussed in the section on Regulatory Issues.

Production of PEF

Very little has been published concerning the manufacture of process engineered fuels. Beyond a few captive operations within the paper and forest products industry, there are probably fewer than twenty commercial PEF producers manufacturing a true PEF product operating in the U.S. today. World-wide, the total production of PEF is believed to be less than 2 million tons. APC believes that advances in technology could expand the use of PEF significantly.

The production of PEF requires specialized equipment and reliable quality control procedures. A representative PEF plant might include the following processing steps:

Specified Feedstock Acquisition→Feedstock QC→First Stage Size Reduction→Separation/Blending→Second Stage Size Reduction→Separation/Blending→Densification (optional)→Product QC

Densified forms of PEF are produced by either a cubing or a pelletizing operation using size reduced feedstocks. Moisture content is an important variable. Table 3 compares representative properties of pellets, cubes, and non-densified PEF. The authors believe that proper size reduction is one of the most critical factors when incorporating higher loadings of plastics. Much more work needs to be done to optimize fuel pellets and cubes containing high levels, i.e., greater than 10% by volume of plastics. Heated dies are being explored to facilitate the incorporation of higher loadings of plastics while maintaining good structural integrity of the densified fuel and associated handling properties.

PEF Economics

The overall economics of PEF are highly case-specific and very dependent of feedstock acquisition costs, disposal avoidance costs, and markets. Reference 10 provides a good introduction to PEF economics. An often overlooked factor is the ability of PEF to compete with other alternative fuels, not

just coal, on a dollars per million Btu delivered basis. All other things being equal, PEF becomes more competitive as its Btu value increases, i.e., plastics content, increases. For a densified PEF of 7500 Btu per pound, a value of approximately \$0.80 per million Btu is representative. At this price, PEF can be competitive with many other alternative fuels including agricultural waste and hog fuel. Any complete financial study must also include the cost to retrofit an industrial or utility boiler to handle PEF in either densified or non-densified form.

One PEF scenario that has been studied at some length is the addition of a PEF production capability to a mixed waste processing facility. ¹¹ A key variable is avoided disposal cost. Capital costs start at about 0.75 million dollars to add densified PEF production capacity to typical mixed waste processing facilities depending on the amount of infrastructure already available such as a building to house the system and the throughput of the facility. Figure 5 shows the relationship between net PEF revenues (fob PEF market) and net savings to the mixed waste processing facility for recovering targeted materials as PEF as compared to landfill disposing of the same materials. The analysis shows that the production of PEF can be cost-effective in some regions of the U.S. In the final analysis, the PEF industry will need to grow and broaden its base of operation before real world economics can be identified and reliably analyzed.

Characterization of PEF

Beginning in late 1995, APC began to characterize samples of commercial and developmental PEFs and to compare their fuel properties with fossil fuels and other alternative fuels. The database presently consists of seventeen samples and continues to grow. Residential, commercial, and industrial feedstocks are included among the samples tested. Representative higher heating value (HHV), chlorine, nitrogen, and sulfur data for a number of samples in the database are shown in Figures 6-9. HHVs ranged from about 5000 Btu/lb to over 15000 Btu/lb. In general, most of the samples easily meet the criteria for PEF presented in Table 2. Sulfur content was consistently low, and over 90 percent of the samples had a chlorine content below 0.5%. The complete database includes ultimate, proximate, ash fusion temperature, ash mineral content, trace metals (17) and TCLP analyses. A detailed report on these findings is being prepared.

PEF Test Burns

Published information on the performance of PEF in industrial and utility boilers is limited. Test burns of densified PEF co-fired with coal in a spreader stoker traveling grate system at Argonne National Laboratories has been reported by Ohlsson. $^{12}\,$ Blends of densified PEF with coal up to 50 percent based on Btu content were investigated. In these experiments the PEF was sourced from a residential and commercial mixed waste processing facility and involved special processing including the addition of lime as a binding agent. The PEF averaged about 7500 Btu/lb. The results showed a reduction in flue gas SO2 , NOx and CO2 when PEF was substituted for high sulfur coal. The same paper reported favorable results co-firing a similar paper derived fuel with coal at the Otter Tail pulverized coal boiler. However, the Btu value of the fuel used at Otter Tail was reported to be only 6400 Btu per pound and therefore does not meet the definition for PEF used in this paper. PEF and RDF did not negatively affect ash quality in any of these tests.

Recently, Doraiswamy, et al., ¹³ reported results from the co-firing of a secondary fiber/plastics recycled paper mill reject stream with coal. This process stream had a heating value above 10,000 Btu/lb and on this basis would be considered PEF. The reject stream contained about 40% plastic on a dry basis. The tests showed no adverse affects on emissions or ash quality co-firing 10% rejects with coal. Economics looked favorable and the co-firing of this type of PEF at other mills and industrial sites can be expected to grow.

Except for some recent Finnish studies reported by Martin Frankenhaeuser, ¹⁴ very little has been published on the co-firing of process engineered fuels in fluidized bed systems. Last year, a two day trial burn of densified PEF derived from commercial paper and plastic feedstocks was completed using the fluidized bed boiler at the Idaho National Engineering Laboratory. The boiler has a maximum rated capacity of 67,500 lb steam/hr.

AUST ARTS CONTRACT

Fuel characterization data for the coal and PEF is shown in Table 4. This was a true PEF with a HHV of 7,449 Btu/lb. The test program measured emissions of trace metals, particulate matter, hydrochloric acid (HCl) and gaseous species (for NO_x, SO₂, CO₂, O₂) while firing three different PEF pellet to coal rations (10%:90%, 20%:80%, 25%:75%) in the boiler. Baseline emissions of the species listed above were also measured while firing only coal in the boiler. The coal was relatively low in sulfur (0.37%) and had a higher heating value of 12,600 Btu/lb. Boiler performance was uncompromised at all pellet feed ratios and emissions profiles held essentially constant even at the highest PEF to coal ratio. Preliminary emissions data for CO₂, NO_x, and SO₂ are shown in Table 5. A complete report of this test burn will be available later this summer. The role of PEF containing sufficient quantities of plastics and the effect of such plastics in achieving HHVs in excess of 10,000 Btu/lb needs to be more thoroughly documented.

In the Fall of 1995, APC completed a bibliography of published information on refuse derived fuels and process engineered fuels including test-burn reports. ¹⁵ The bibliography is available from APC in hard copy and disk format.

Regulatory Issues

Regulatory Issues were prominent on the agenda during the recent Pellet Fuel Conference held in Washington, DC. ¹⁶ Regulations, laws, and policies can act as drivers or barriers to the use of PEF as a supplemental fuel. Regulatory issues affecting the use of PEF are briefly discussed below and fall into four categories.

- Definitions and terminology
- Fuel standards and quality assurance
- Air emissions standards
- Ash management

PEF Definitions and Terminology. The concept of PEF is relatively new, so federal and state regulations generally do not contain a definition for PEF. Regulators may be inclined to interchangeably use the terms "PEF" and "RDF", but the general consensus of the industry is that PEF is higher quality and more homogeneous fuel than RDF, as noted earlier. In order to minimize barriers to the growth of PEF use, it is important that PEF be considered a resource by regulators rather than a waste, in order to avoid the generally more stringent regulations associated with waste management as compared to resource management. Earlier, this issue also had to be addressed by the traditional recycling industry in order for it to grow to its current level.

PEF can also be considered an indigenous and renewable fuel because it is generally composed of postuse paper and plastics. Clearly defining PEF as a renewable fuel would further the industry by allowing PEF to qualify for incentives or mandates established to encourage the use of renewable fuels in the United States, as well as further its perception as a valuable energy resource. This issue is developing as an important consideration during the electricity deregulation discussions underway at both the state and federal levels.

Some states offer recycling (or waste diversion) credit for materials that are diverted from disposal through PEF systems, thus acknowledging their role in resource conservation. For example, Florida and Maine provide recycling credit to local governments for combustion of PEF in existing boilers. It is interesting to note that recycling credit in Florida cannot be obtained for combustion of the same materials in traditional WTE facilities.

PEF Standards and Quality Assurance. Also because the concept of PEF is still relatively new in the United States, standards and quality assurance measures for PEF have not yet been developed, although some efforts to start this process are underway. An effort to develop similar standards is currently active in Europe for recovered fuel. Ideally, the development of such specifications in the United States would potentially:

- Minimize the stringency of regulations imposed on PEF production and use (such as reducing
 expensive air emissions and ash testing requirements often imposed by regulators on boiler
 operators wanting to co-fire alternative fuels with other conventional fuels) or possibly exempt
 PEF from some regulations;
- Simplify permitting procedures and requirements associated with the production and use of PEF and make them comparable to those for conventional fuels; and
- Facilitate the perception and definition of PEF as a resource and renewable fuel.

The development of standards and quality assurance for PEF must address both producer and user responsibilities and specific sampling and testing protocols. A classification system based on key properties is a possibility.

Air Standards. Under the Clean Air Act, up to 30 percent by weight of MSW can be co-combusted with other fuels without triggering municipal waste combustor (MWC) regulations. It is probable that PEF, with one or more source separated types of post-consumer materials such as paper, would fall under the 30 percent restriction. However, PEF made from industrial scrap that has not been commingled with MSW may not fall under the same percentage restriction. At the state level, additional restrictions are sometimes placed on the amount of PEF that can be co-fired before triggering MWC regulations. For example, Washington state has a limit on the use of PEF that supersedes and is more stringent than the 30 percent exemption in the Clean Air Act.

Regulations that govern WTE facilities, or MWC regulations, are generally more stringent for certain pollutants (such as organics and trace metals) than regulations for boilers fired with conventional fuels. Therefore, it is possible that co-firing PEF in existing boilers will be less stringently regulated than combusting MSW in WTE facilities. However, regulatory consequences could result from co-firing PEF depending on the emissions consequences. Testing will most likely be required to determine the impact of co-firing PEF on emissions unless strict PEF standards and quality assurance measures are developed.

Co-firing of PEF with conventional fuels, particularly coal, can have positive affects on air emissions. Most notably, co-firing of PEF with coal offers a potentially more cost-effective means to achieve compliance with the federal Acid Rain Program of the 1990 Clean Air Act Amendments than through installation of scrubbers or other capital-intensive alternative compliance strategies.

Anticipated regulatory changes could affect the way and the amount of PEF used by industrial combustion facilities. It is anticipated that relevant definitions and regulations will be thoroughly revisited as part of the Industrial Combustor Coordinated Rulemaking Process underway at the U.S. Environmental Protection Agency. The uncertainty regarding environmental regulation of PEF use can be a barrier to the development of the PEF industry. Some state-level restrictions on the amount that can be co-fired before triggering MWC regulation can also be barriers, as well as costs for ongoing required testing can be prohibitive to the use of PEF. However, potential positive effects on air emissions are helping to drive the industry.

Ash Management. The regulation of ash resulting from the co-firing of PEF with conventional fuels is not clear. Currently, ash from combustion of coal is treated as a "Bevill waste" and need not be tested for hazardous characteristics. Bevill waste can be fired with up to 50 percent of other types of fuels and not be subject to Subtitle C regulation. However, the Clean Air Act exemption of 30 percent does not clearly exempt the resulting ash from MWC ash regulations which includes testing requirements for hazardous characteristics. Also, it is not clear if ash from boilers co-firing PEF from residential sources is exempt from Subtitle D regulations and there is no clear guidance on the federal level regarding how ash from a facility co-firing PEF derived from industrial feedstocks should be regulated. Federal legislation or regulatory clarification of these issues could improve the use of PEF.

States generally have the primary responsibility for regulating the management of ash from facilities cofiring PEF although general guidance is typically provided from the federal government such as that discussed above. Typically states require ash from facilities co-firing MSW with any type of conventional fuels in amounts greater than 30 percent of the total fuel to be regulated the same as ash from MWC's. Ash from co-firing PEF derived from industrial feedstocks with coal at levels greater than 50 percent must generally be tested for hazardous characteristics. Regulations for management of ash from co-firing wood waste with PEF derived from industrial sources at any percentage must be completely determined at the state level since no specific federal regulations address this issue.

In general, competition in the fuel and energy industries is increasing as a result of in place and planned deregulation of the electric utility industry. Increased competition is resulting in a reassessment of alternative fuels. PEF, on the basis of overall environmental performance and economics, has the potential to be the alternative fuel of choice in many situations.

REFERENCES

- 1. Characterization of Municipal Solid Waste in the United States: 1995 Update, EPA530-R-96-001, March 1996.
- 2. The 1996 IWSA Municipal Waste Combustion Directory of the United States Facilities, Integrated Waste Services Association, Washington, DC, 1996.
- 3. *PEF Economic/Business Viability Analyses Report*, R.W. Beck & Associates, (APC report) January 1997.
- 4. F. Mark, Energy Recovery Through Co-Combustion of Mixed Plastics Waste and Municipal Solid Waste, APME Technical Paper, Brussels, June 1994.
- 5. The Relationship Between Chlorine in Waste Streams and Dioxin Emissions from Waste Combustor Stacks, ASME Research Report, CRTD-Vol. 36, 1995.
- 6. An International Perspective on Characterisation and Management of Residues from Municipal Solid Waste Incineration--Summary Report, The International Ash Working Group, July 1995.
- 7. Energy Recovery from Automotive Shredder Residue--Summary Report, Roy F. Weston, Inc., May 1995.
- 8. G. Sutherland and L. Tomczyk, *Pellet Fuels*, <u>Proceedings of The International Conference on Solid Waste Management, Thermal Treatment & Waste-to-Energy Technology</u>, Air & Waste Management Association, Washington, DC, 1995, pp 135-146.
- 9. A Fuel for the Future: Energy from Plastics Waste, Association of Plastics Manufacturing in Europe, Brussels, Belgium.
- 10. L. Tomczyk, Engineered Fuel: Renewable Fuel of the Future? Solid Waste Technologies, January/February: 25 (1977).
- 11. Processed Engineered Fuel Economic Case Study, R. W. Beck & Associates, (APC report) June 1996.
- 12. O. Ohlsson, Development and Evaluation of Lime Enhanced Refuse-Derived Fuel (RDF) Pellets, Proceedings of The International Conference on Solid Waste Management, Thermal Treatment & Waste-to-Energy Technology, Air & Waste Management Association, Washington, DC, 1995, pp 124-134.
- 13. J. Doraiswamy, N. Sell, J. Norman, G. Massey, K. Nelson, and M. Doshi, Use of Secondary-Fiber Rejects as Fuel for a Coal-Fired Stoker Boiler, <u>TAPPI Journal</u>, 79(3), 199 (1996).

- 14. M. Frankenhaeuser, H. Manninen, I. Kojo, J. Ruuskanen, T. Vartiainen, R. Vesterinen, and J. Virkki, *Organic Emissions from Co-Combustion of Mixed Plastics with Coal in a Bubbling Fluidized Bed Boiler*, Chemosphere, Vol. 27, Nos. 1-3, pp 309-316, (1993).
- 15. Annotated Bibliography of Refuse Derived Fuel (RDF) References and Densified Refuse Derived Fuel (dRDF) Pellet Test Firings, Argonne National Laboratory, November 1995.
- 16. Proceedings, Pellet Fuels Conference, November 16-17, 1995, Washington, DC.

Table 1. Municipal Waste Combustion Industry Profile (1995)

	Number	Annual Operating Capacity (MM tons)
Mass Burn Facilities	70	22.6
RDF Combustion Facilities	23	7.9
Modular Facilities	21	0.8
Incinerators (without energy recovery)	21	0.9
States with Facilities	34	
Communities Served	1688	
Population Served (million)	41.2	
Total Capacity		32.2
Percentage of MSW Processed	15.4	

Source: Adapted from The 1996 IWSA Municipal Waste Combustion Directory

Table 2. Comparative Properties of Characteristics of PEF, RDF, and MSW

Type of Fuel	Moisture (% by wt)	HHV (Btu/lb)	Ash Content (% by wt)	Sulfur Content (% by wt)	Chlorine Content (% by wt)
MSW, unprocessed	15-50	4,500-5,500	18-30	0.10-0.50	0.10-1.00
RDF	3-35	5,500-6,500	8-25	0.10-0.50	0.10-1.00
PEF	3-20	6,500-16,000	2-15	0.02-0.20	0.03-0.50
Bituminous Coal	2-20	11,000-14,500	3-16	0.50-4.7	0.01-0.90

Table 3. Comparison of Pelletizing and Cubing Technologies

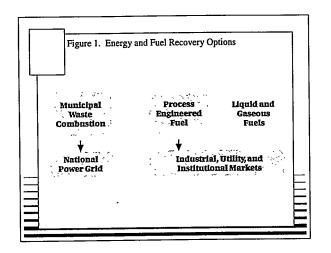
Criteria	Pellets	Cubes	Non-densified
Size Reduction Required	1/4" to 3/4" max size	3/4" to 3-1/2 (1-1/2" nominal)	1/4" - 2"
Final Bulk Density	35-45 lbs/cu ft	20-33 lbs/cu ft	2-12 lbs/cu ft
Final Product Diameter	1/4" to 3/4"	1" to 1-1/4"	N/A
Cooling Required After Densification	Yes	No	N/A
Throughput	2-4 tons per hour	6-8 tons per hour	> 5 tons per hour

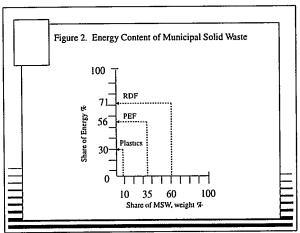
Table 4. Fuel Characterization - INEL Test Burn

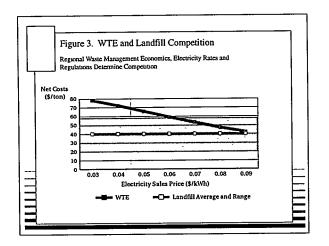
Fuel Type	C	H	O	N	S	HHV
	(%)	(%)	(%)	(%)	(%)	(Btu/lb)
Coal	73.33	4.78	10.75	1.20	0.37	12,582
PEF	43.31	5.74	35.40	0.32	0.18	7,449

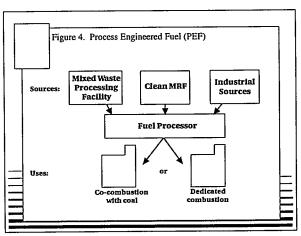
Table 5. INEL emissions data.

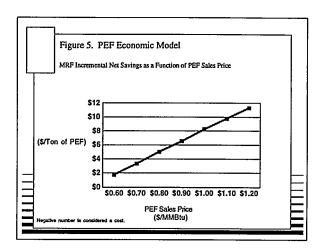
Coal/PEF Ratio	100%/0%	90%/10%	80%/20%	75%/25%
CO ₂ , %	9.58	9.60	9.57	9.00
NO _x , ppm @ 7% O ₂	239	210	197	212
SO ₂ , ppm @ 7% O ₂	29	34	43	45

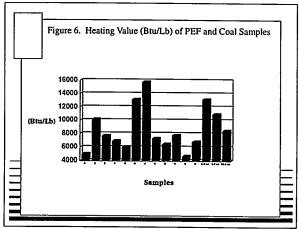


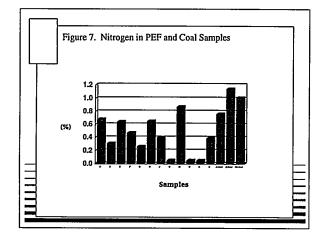


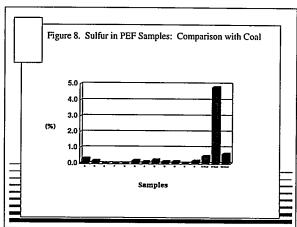


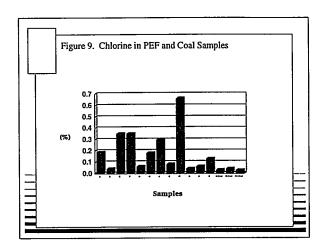












EPA's Mercury Report To Congress: A Basis for Decision-Making?

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INTRODUCTION

Section 112 (n)(1)(B) of the Clean Air Act Amendments of 1990 directed the US EPA to conduct and transmit to Congress by November 1994, a study of mercury emissions from electric utility steam generating units, municipal waste combustion (MWC) units, and other sources including area sources. The study was to consider the rate and mass of the emissions, the health and environmental effects of these emissions, technologies to control them, and the costs of the controls.

While the Report was intended to inform Congress concerning mercury emissions, it is also intended to serve as a basis for future Federal policy and regulatory decisions. The purpose of this paper is to review and assess the status of the Report and identify its weaknesses and deficiencies. It is the studied opinion of this author that the Report, in its current format, suffers from a number of "fatal flaws" which bring into question its usefulness both as an accurate information document for Congressional review, and as a basis for future policy and regulatory decisions.

The discussion below begins with a brief look at the history of the Report. Specific sections of the Report are identified where perhaps fatal flaws exist in the areas of sources, fate and transport models, and some of the controversy over mercury exposure, health effects, and the chosen reference dose. Some observations are incorporated concerning the Science Advisory Board's comments to EPA in a recent review panel. Finally, these comments address flaws in the current Report, but also identify and address flaws in the EPA Report preparation process.

MERCURY REPORT TO CONGRESS

History

For a number of years beginning around 1987, representatives of EPA's Offices of Air Quality Planning & Standards (OAQPS) and Research and Development (ORD) had been involved with collecting data on mercury emissions from MWCs under the Clean Air Act Section 111 New Source Performance Standards development activities. In gathering emissions data, these EPA representatives interacted with MWC owners and operators individually and through groups such as the Integrated Waste Services Association (the MWC industry trade association), the Conference of Mayors/Municipal Waste Management Association (MWMA), Solid Waste Association of North America (SWANA), and the American Society of Mechanical Engineers (ASME).

As part of this continuing effort and dialog, OAQPS held a two-day Public Meeting in February 1990, at their offices at Research Triangle Park, NC to review the status of their NSPS data collection activities for mercury from MWCs. This information and subsequent data eventually lead to promulgation of an emissions standard for new and existing MWCs (40 CFR 60.50b and 60.30b) in December 1995.

After the passage of the 1990 CAAA, EPA's OAQPS's Emission Factor and Inventory Group also began collecting various emissions information to prepare a series of reports on pollutant emissions, e.g., Locating and Estimating Air Emissions from Sources of Mercury and Mercury Compounds. However, the individuals within this branch of OAQPS were not the same ones as those working on the MWC NSPS.

The MWC industry first became aware of EPA's work in preparing the Locating and Estimating Report in June 1993. The Integrated Waste Services Association (IWSA) provided comments to EPA on MWC

sources and emission rates of mercury. In September 1993, EPA issued the Locating and Estimating Mercury report, ignoring the majority of IWSA's work. Three months later, EPA issued a draft report entitled: National Emissions Inventory of Mercury and Mercury Compounds: Interim Final Report and solicited comments on selected sections of it. IWSA responded by referring the Agency to its previous detailed comments to the Locating and Estimating report.

Over the next year, there was no further communication from EPA on the Mercury reports. Then, in January 1995, EPA published a notice in the Federal Register that there would be a two-day Peer Review workshop to discuss the results of the draft Mercury Report to Congress. While the workshop was open to the public, EPA was not inclined to accept any public comments.

At the January meeting, it was learned that EPA had proceeded with the development of the Mercury Report to Congress, and while one of the major stakeholders, the electric utility industry, had been integrally involved in the preparation and review of the Report, the MWC industry was not even informed of the existence of a draft report until the Public Notice. In addition, no one from EPA's OAQPS or ORD that had been working for the past several years on the MWC NSPS and data collection had been involved in the preparation or review of the draft Report.

The draft Report was provided to the public, including IWSA, for the first time on the morning of the first day of the Workshop. The Public was asked to review and comment on the seven volume draft report on the same day they received it. IWSA was granted only an "observer" status and was offered ten minutes to present any comments at the Workshop.

While EPA was not inclined to accept any public comments on the draft Report, IWSA was subsequently allowed to submit a comment document which cited factual errors in data as well as errors and problems with many of EPA's assumptions in the report. One of the most fundamental flaws in the draft report was that the basic emissions data in the report had been drawn from the incorrect Locating and Estimating - Mercury Report. EPA had ignored the data submitted by IWSA, some of which was based upon testing conducted by EPA's OAQPS and ORD.

Problems with the Draft Report

The draft Report contained inaccuracies and erroneous and ultraconservative assumptions which could lead a reader to believe that mercury emissions from MWCs were seriously impacting the environment and threatening human health. The three primary problem areas in the draft Report are: 1)Sources of mercury emissions, 2) Fate and Transport Models and Assumptions, and 3) Toxicologic information and assumptions.

Sources. As stated above, the branch of EPA that collected emissions data for the draft Mercury Report relied on old data. All of the actual stack test results of mercury emissions from MWCs that had been generated for State air permit compliance tests and by EPA/ORD and OAQPS for development of the MWC NSPS was ignored. These data were readily available and much of it had been provided to EPA by IWSA in comments on the Locating & Estimating and National Inventory reports.

Also, even though the draft report was released in January 1995, EPA based the source data in the Report on data as it existed in 1990. In the draft Mercury Report, EPA's estimate of mercury emissions from MWCs was 64 tons per year (tpy), even though in 1993, IWSA provided EPA with data demonstrating that 1993 emissions were no more than 44 tpy and that value was decreasing rapidly. The Agency's own most recent inventory estimate for MWCs for 1995 is 29 tpy.

The 1995 draft Mercury Report states that MWCs account for 12% of the emissions inventory. However, in 1995, EPA issued a Fact Sheet along with the release of the MACT Emissions Standards for MWCs which stated that MWCs will represent only 3% of the total mercury emissions nationwide once the MACT standards are fully implemented. The inference here is that current MWC emissions are four times what they will be by December 2000. This is patently untrue as is clearly shown in the MWC emissions database that is maintained by EPA.

EPA also ignored other sources of mercury emissions, references for which were readily available in reports prepared by states such as New Jersey and Florida, and underestimated other sources even when they did include them. For example, EPA underestimated the potential mercury releases from fluorescent lights by an order of magnitude, totally ignored the potentially large source of mercury emissions from open field burning, and used inaccurate mercury speciation data in characterizing emissions from MWCs.

Fate and Transport Models and Assumptions. In the draft Mercury Report, EPA relied on series of equations and models to estimate the potential impact of emissions of mercury from U.S. sources. The two air quality models were a regional prediction model and a source specific model. The rudimentary multiple pathway model, IEM2, which is primarily a series of equations, was also used. Comments on the inadequacies of these models and the problems with the associated assumptions made by EPA were provided to EPA by IWSA. The following are some of the more critical points made by IWSA.

RELMAP. To predict regional concentrations of mercury from sources of mercury emissions, EPA used the Regional Lagrangian Model of Air Pollution (RELMAP), and attempted to verify the results of the RELMAP predictions by comparing them to specific observations and sampling events. However, the actual measured results prove the RELMAP-predicted values consistently overestimate actual wet deposition data by a factor of two. EPA states in the Report that actual results "agree well" with predictions, even though EPA's data proves this to be incorrect. For example, EPA states in the Report that at one location in Vermont, the observed measurements were 9.3 ug/m², whereas the RELMAP model predicted up to 20 ug/m². For another location, the RELMAP model predicted deposition at up to 10 ug/m² when actual deposition was 5.7 ug/m².

The RELMAP model predicted local maximum values for mercury wet deposition as 60 ug/m², while EPA cites the highest measured value of actual data as 30 ug/m² in the Florida Everglades. As shown by the data in EPA's report, predicted impacts using RELMAP are approximately twice the actual measurements in every case. In the Report, the Agency even admits that the RELMAP results were "significantly larger" than the measured values. However, no attempt to adjust the RELMAP results were made.

While a factor of two may not seem large enough to be of concern, the impact of the RELMAP predicted values is significant on the overall assessment of potential risk. In EPA's hypothetical risk assessment, RELMAP results contribute to a mercury concentration in fish of 0.5 ppm, *one-half* the FDA action level. Consequently, a factor of two becomes very significant, dropping the regional contribution to 0.25 ppm in fish.

COMPMERC. To estimate the impact from specific sources of mercury emissions, EPA used COMPMERC, a modified version of a dispersion and deposition model called COMPDEP. Air quality models are usually developed for or by OAQPS's Air Modeling branch in Research Triangle Park, NC (RTP). The COMPDEP model, however, was developed by EPA's Environmental Assessment office in

Washington, D.C. The DC office, recognizing that COMPDEP had not been validated nor peer reviewed asked the RTP modeling group to refine COMPDEP. The RTP office had been developing and was about to promulgate the next version of their dispersion model, ISC3, which had been peer reviewed and was more sophisticated than COMPDEP. The RTP group was unable to refine nor validate the wet deposition algorithms in COMPDEP and, in an effort to respond to the request from the Environmental Assessment group, the Modeling group added the wet deposition portion of COMPDEP to the ISC3 model which also included a dry deposition component. The ISC3 model with the COMPDEP wet deposition algorithms was released to the public by EPA along with the caveat that anyone using the wet deposition portion of ISC3 does so "at their own risk" and that the wet deposition portion of ISC3 "should not be used for regulatory purposes", e.g., risk assessment or permitting (Personal communication, OAQPS, Sept. 1996).

It is puzzling as to why EPA's Environmental Assessment group chose to use COMPDEP for the Mercury Report. COMPDEP suffers from many flaws, such as lack of source depletion and poor handling of wet and dry deposition. EPA's Science Advisory Board criticized EPA in February 1996 for use of unvalidated modeling, including COMPDEP and IEM2, saying that the models "violate a fundamental scientific principal, i.e., conservation of mass" and provide "little grounding of reality of the overall methodology."

While EPA's newer model, ISC3, is not a perfect model, it contains a number of improvements over the COMPDEP model, including a plume depletion algorithm designed to conserve mass. Unfortunately, the wet deposition algorithms in ISC3 are the COMPDEP algorithms that have still not received the validation necessary to allow its use as a regulatory tool.

The wet deposition portion of COMPDEP is seriously flawed. For modeling of wet deposition, COMPDEP estimates the maximum wet flux rate and assumes that this occurs at the same point as the maximum dry deposition. It is well known that wet deposition flux rates drop off quickly with distance from the stack, and maximum impacts typically occur within several hundred meters of the stack. The maximum dry deposition impact point, however, occurs much further away from the stack. The wet flux rates can decrease by more than an order of magnitude within a few kilometers of the maximum point. The maximum wet flux rate may be more than 100 times higher than the average flux rate across a 10-km modeled area. Use of the maximum impact point wet flux rate will overestimate long term risks by up to two orders of magnitude.

Air Quality Model Input Parameter Assumptions. EPA also made many assumptions in using the RELMAP and COMPDEP models. The base-case mercury speciation profiles for MWCs were contradictory and without reference. For the RELMAP modeling of MWCs, one table in the Report indicated a base case speciation of 15% elemental, 85% ionic, and 20% particle bound, while another table had the base-case speciation for the same source listed as 20% elemental, 60% ionic, and, 20% particle bound. For the COMPDEP modeling of MWCs, the speciation was assumed to be 15% elemental, 85% ionic, with no particle bound mercury. Thus, three different base-case speciations are listed for the same plant type, all without reference.

The mercury speciation assumed in the draft Report of 15% elemental and 85% ionic for MWCs is also inconsistent with other published data. According to work completed in 1991 by Dr. Nicolas Bloom at the Stanislaus, California waste-to-energy plant, a more accurate percentage is 70% elemental and 30% ionic for an MWC with a scrubber and fabric filter baghouse.

Within the next two to three years, most MWCs will be injecting powdered activated carbon into the flue gas as a mercury emission control method to comply with the 1995 MACT standard. Results of testing conducted by EPA and others show that powdered activated carbon is extremely effective in capturing ionic mercury. Actual results using carbon to control mercury emissions demonstrate at least 90% removal of mercury from MWC flue gases. After MACT controls reduce total mercury emission rates by 90% or greater, the remaining species will be between 70%-90% elemental mercury and between 10%-30% ionic mercury. Therefore, the use of elemental to ionic ratios of less than 70:30 is misleading.

EPA did acknowledge that "due to the high degree of uncertainty regarding the emission speciation and possible rapid chemical and physical transformations immediately after emission, it is recommended that these maximum simulated deposition values should be considered highly uncertain until further research is conducted to reduce these uncertainties." Despite this acknowledged "high degree of uncertainty," EPA continued to use speciation values without reference when, in fact, actual field data is available.

Using unvalidated air models and models that consistently over predict along with invalid assumptions can yield projected impact results from specific sources that are orders of magnitude higher than they really are.

Water Quality and Terrestrial Models. The model used to predict concentrations in the aquatic and terrestrial compartments, the Indirect Exposure Methodology (IEM2) model, is based on a series of equations initially developed for organic chemicals and is not directly transferable to inorganic mercury compounds. In addition, there are errors in the equations that were used in IEM2. For example, the equation used to predict the gas phase mass transfer coefficient was derived for surface spills of organic liquids and is determined based on the area of the spilled material. This equation is not appropriate for mercury, especially in the case of an ionic species such as Hg(II) or monomethylmercury.

The diffusion of mercury from air to the ground surface or water should be governed by a concentration gradient, but the IEM2 model does not address this factor. The IEM2 model equations also do not address any chemical transformations affecting mercury in soil, water or sediments (oxidation, reduction, methylation, demethylation), nor do they address physicochemical properties important to inorganic chemicals (complex equilibrium, dissolution, precipitation). The chemistry of mercury is complex and dynamic and must be accounted for if a reasonably accurate picture is to be developed concerning mercury's behavior in the environment.

EPA also notes that the model "cannot be used to predict response to reduction or elimination of loadings." Thus, the IEM2 model treats lakes like a bathtub without a drain. If a source reduces its emission or stops emitting altogether, there is no decrease in deposition over time. This is an obvious critical flaw in the model. This disconnect between reality and the IEM2 modeling methodology results in an inaccurate depiction of the potential impacts of mercury.

In addition, the models used to predict sediment concentrations have not been validated, and are likely to produce unrealistic results. Based upon the above, it is estimated that EPA's use of IEM2 and the associated assumptions result in a one to two orders of magnitude overestimation of mercury impacts.

Impacts on Fish. The methodology used to calculate concentrations of mercury in water does not adequately address the complex nature of the mercury biogeochemical cycle in the aquatic environment. The EPA model structure is overly simplified, with the soil-water partition coefficient assuming

inordinate importance in predicting mercury behavior. Many other equally or more important factors and reactions affecting mercury behavior in reality are not addressed in the model, including acid-base reactions, oxidation-reduction reactions, methylation, complexation, water temperature, pH and dissolved organic carbon. Models that incorporate the factors affecting the behavior of mercury are important adjuncts to predicting bioaccumulation. EPA, however, rejected the use of more refined approaches such as the mercury cycling model (MCM) outlined in Hudson et al. (1994), choosing instead "to accept that considerable variation exists in mercury bioaccumulation in fish". The result of these decisions is a low degree of confidence in the results from the IEM2 model. Numerous alternative aquatic models (e.g., REDEOL, WATE02, MINEOL, MEXAMS) exist that can be used to deal with the equilibria of mercury in aquatic systems. These models incorporate pH effects, redox chemistry, precipitation, surface adsorption, ligand exchange and formation of ternary complexes.

The methodology that EPA used to address mercury bioaccumulation in fish also suffers from uncertainty. The Agency notes in the draft Report that there is a "substantial level of uncertainty" in the bioaccumulation values for the two trophic levels of fish studied, and shows that both the derived bioaccumulation factor values and the bioaccumulation factor values based on estimates derived from the literature vary by *more than two orders of magnitude*. The Agency nevertheless uses point estimates for bioaccumulation factors to predict deterministic exposures in the Report without ever analyzing the precise level of uncertainty.

CONGRESS and FEDERAL AGENCIES

Voicing concern over the conclusions in the draft Report, several federal agencies, including the Food and Drug Administration and the Departments of Agriculture, Commerce and Energy, requested that they be allowed to review the Report before it was released (Environment Reporter, April 1996).

One of the primary concerns expressed was how little mention there was in the Report of the National Institute of Environmental Health Sciences (NIEHS)-funded study of the Seychelles Islands. This comprehensive multi-year study was an attempt to overcome the problems of all of the other epidemiologic studies of the health impacts of mercury exposure. EPA based its human health impact analyses in large part upon the results of the study of the mercury contaminated grain episode in Iraq in 1971. The Iraqi study, which was after-the-fact and anecdotal, indicated that fetal exposure to very low levels of mercury would lead to developmental problems. However, many researchers, including the researchers that conducted the Iraqi study have cautioned that the Iraqi study should not be used to derive toxicological assumptions for mercury. EPA also reviewed other mercury-related human health studies, but in most of these case studies, there were limitations and/or confounding factors.

The Seychelles' study is being conducted by a team from the University of Rochester and includes members of the team that conducted the Iraqi study. According to NIEHS, the Seychelles Islands study, "a study of children of heavy, daily fish consumers, demonstrated no developmental damage from the same levels of mercury exposure that was found in the Iraqi study." (NIEHS Press Release, 1996.)

Subsequent to the release of the 1995 draft Report, EPA did make some changes in the Report which did address some of IWSA's comments. For example, EPA included three mercury emission speciation scenarios for MWCs: the original speciation assumption and two assumptions which more closely relate to actual measured data from MWCs. The modeling results using these alternative scenarios, which showed insignificant impacts, were not discussed in the Executive Summary nor the Summary and

Conclusions of the revised Report. These speciation scenarios are found on one page of text and three tables in the middle of the 100-plus page Volume III of the revised draft Report.

SCIENCE ADVISORY BOARD

Concerned that EPA might complete the Mercury Report before the important Seychelles work was published, a bipartisan group of Senators and members of Congress asked EPA to delay finalizing the Report until the Seychelles study was released and could be incorporated into the EPA Report. This group also asked EPA to submit the draft Report to EPA's Science Advisory Board (SAB) for review. On June 18, 1996, EPA announced that it was delaying the release of the Report and had sent the revised draft Mercury Report to the SAB to begin its review.

On February 26 and 27, 1997, the Mercury Subcommittee of the SAB held a Public Meeting to review their findings and receive comments from the public on the draft Report. According to published accounts (Daily Environment Report, February 1997) the Subcommittee strongly criticized the computer risk assessment models used by EPA as inadequate and unable to predict impacts from specific sources, and that EPA should not imply that the models are capable of accurately predicting impacts on humans, wildlife or aquatic life. The SAB panel pointed to EPA's own admission that the models are incapable of predicting the change in impacts on human health or wildlife due to decreases in any particular source.

The panel also reiterated EPA's own statements that the report and the estimates of risk are based on **qualitative** and not **quantitative** analyses. The panel suggested that EPA either remove the predictive models altogether from the report or make improvements and rerun the models with the qualifier that the results are still qualitative and should not be used for regulatory or risk assessment purposes.

The Subcommittee also pointed out that there is a large amount of mercury in the environment today that is the result of historic practices as well as ongoing activities such as gold and copper mining in South America and Asia that the U.S. has little ability to control. As this reservoir of mercury is re-emitted into the environment, it may equal the total current anthropogenic emissions. Therefore, additional controls on current mercury emissions in the U.S. may have little impact on the amount of mercury deposited in the U.S.

The Subcommittee nonetheless endorsed the Report's overall conclusion that exposure to methylmercury is harmful even at relatively low doses based upon the Iraqi and other studies cited in the draft Report. The Seychelles' studies included a small number of outlier results which the Seychelles' researchers felt could justifiably be removed from the evaluation. Without these outliers, the Seychelles' study showed no health impacts at the low levels reported in the previous studies. With the outliers, the results were unclear. The Panel believed that there was insufficient justification for discarding the outliers, and that meaningful results from Seychelles studies were still two years off.

The Panel found that EPA's current reference dose (Rfd), based on the Iraqi study, was within an order of magnitude of that suggested the other studies EPA reviewed and the Seychelles' study, and that this is within an acceptable margin of safety. Therefore, the Panel determined that EPA should not revise its Rfd.

SUMMARY

There are so many errors and questionable assumptions in the draft Mercury Report to Congress that it would take many more pages to discuss them all in detail than can be done so here. Some of the errors don't even pass a logic test, e.g., in the Report, EPA predicted that a single large municipal waste combustor could cause a mercury concentration of 26 ug/g in fish in a nearby lake, and yet, the highest concentration of mercury in fish in EPA's database is 8.9 ug/g. No fish sampled near an operating MWC have ever yielded a mercury level of even approaching 8 ug/g.

In many parts of the report, EPA states that there is substantial uncertainty or variability inherent in the modeling input parameters, such as the deposition flux rate or the fish bioaccumulation factor and fish ingestion rates. However, these caveats are not adequately carried through to the results of the analysis and therefore do not allow the reader to place the information into a reasonable context.

EPA has taken the approach of speculating on the worst case, hypothetical, potential impact that might ever occur using the most rudimentary and convenient models and assumptions. The failure to carry acknowledged uncertainties through into the main body of the analysis will lead to misinterpretation of results. These errors and assumptions can result in overestimating predictions of potential risk by several orders of magnitude.

The EPA Science Advisory Board Mercury Subcommittee has criticized EPA for many of the same reasons that others have, e.g., that characterizing the risk to human health and wildlife cannot be done in a quantitative fashion since the various models do not have the ability to accurately predict the health impacts of anthropogenic source emissions and their indirect exposure pathways. However, the SAB did not suggest that EPA increase the Rfd based upon the results of the Seychelles study. The studies that EPA relied upon were flawed and contain confounding factors which make it difficult to specifically attribute a cause/effect relationship to mercury. Even though the results of the Seychelles study show that the Rfd is within an order of magnitude of the one proposed by EPA, the Rfd for mercury should still be increased since it is based upon a series of studies with questionable results.

Nevertheless, EPA now must address the errors in the Report before a final version is released to Congress and the public.

REFERENCES

Bloom, N.S., "Mercury Speciation in Flue Gases: Overcoming the Analytical Difficulties," Brooks Rand Ltd., Seattle, WA, Fall 1991

Guentzel, J.L, Landing, W.M., Gill, G.A. and Pollman, C.D. 1995. Atmospheric deposition of mercury in Florida: The FAMS Project (1992-1994), Water, Air and Soil Poll. 80:393-402.

Gill, G.A., Guentzel, J.L., Landgin, W.M. and Pollman, C.D. 1995. Total gaseous mercury measurements in Florida: The FAMS Project (1992-1994). Water, Air and Soil Poll. 80:235-244.

Hudson, R,J.M., Gherini, S.A., Watras, C.J. and Porcella, D.B. 1994. Modeling the biogeochemical cycling of mercury in lakes: The mercury cycling model (MCM) and its application to the MTL study lakes. In Mercury Pollution, Integration and Synthesis, C.J. Watras and J.W. Huckabee (Eds.). Lewis Publishers: Boca Raton, FL.

Pollman, C.D., Gill. G., Guentzel, J.L., Bare, D., Porcella, D., Zillioux, E. and Atkeson, T, 1995, Overview of the Florida Atmospheric Mercury Study (FAMS). Water, Air and Soil Poll. 80:285-290.

Reardon, E.J. 1981. K_d s -- Can they be used to describe reversible ion sorption reactions in contaminant migration? Groundwater 19:279-286.

"Release of Mercury Study Delayed by EPA Following Concerns About Health Data Used", Environment Reporter, April 12, 1996, pg. 2333-2335

"SAB Panel Endorses EPA Mercury Report, Recommends Improved Modeling Plan", Environment Reporter, March 7, 1997

"SAB Praises Mercury Report, with Source Modeling Caveats" Risk Policy Report, March 21, 1997.

"Science Advisory Board Panel Reviews Controversial EPA Report on Mercury", Daily Environment Report, February 27, 1997

Trapp, S. and McFarlane, J.C., Eds. 1995, Plant Contamination: Modeling and Simulation of Organic Chemical Processes. CRC Press Inc.: Boca Raton, Florida.

Should Regional Municipal Solid Waste Consortia Enter Incentive Based Service Contracts for Professional Management Services?

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INTRODUCTION

There are myriad challenges facing any regional municipal solid waste management consortium today. Consortia of the sort discussed here include any regional municipal waste authority, board, committee or other governing body. The challenges are especially acute when a waste-to-energy facility is part of the integrated solid waste system. With increasing challenges in the regulatory, technical and financial arenas as well as over-worked staffs, managers of many consortia are turning to professional consultants for a variety of support services. Today, these include, not only the more traditional services, such as engineering, accounting and legal, but also often include business management services, as well.

There are many areas in which solid waste management consultants are asked to provide specialized support to public, waste-to-energy consortia. Three commonly sought services are: a) negotiating contracts and other potentially adversarial matters, b) procuring specialized goods and services and c) marketing facility capacity. In each of these areas, taxpayer dollars can be saved or squandered. Some argue that "success based" or incentivized contracts provide management consultants very strong motivation to get the best deal possible for the client. Remuneration is based on pre-determined benchmarks, and escalates as the client's benefit increases. In some cases, management consultants charge lower than standard rates during a given project, in return for a larger total return if successful.

When appropriate, these contracts can provide excellent results. However, there are circumstances when such arrangements can lead to unexpected, negative consequences. This is particularly true when the client consists of many autonomous municipalities. This paper reviews typical, support services a waste-to-energy consortium might seek from a management consultant. It also discusses incentive based contracts and how to avoid their potential problems.

TYPICAL MANAGEMENT SERVICES

Negotiation Services

Consortium Formation -- Contractual relationships among municipal consortium members, typically include governing, as well as, operational by-laws. These by-laws need to be developed and executed to form the group in the first place. The appropriate regional relationships can, and do, vary dramatically. Some consortia are relatively informal, ad hoc, committees or commissions, focusing upon relatively narrow goals. At the other end of the spectrum are formal, solid waste districts with powers to take land, develop significant facilities and assess taxes. Management consultants, with appropriate experience, can help negotiate agreements that will provide the structure required to accomplish shared goals, without adding unneeded bureaucracy. The significance of a consortium's by-laws, with respect to incentive based contracts, is discussed below.

Contracts – Contracts, for goods and services, between a waste-to-energy consortium and a wide variety of vendors, typically represent millions of dollars. Initial agreements must be negotiated aggressively, in the consortium's interest. After being executed, conditions frequently change, requiring re-negotiation or dispute resolution. The highly specialized nature of many of these goods and services has led some consortia to seek focused assistance to negotiate such contracts. Such service can help develop

appropriate specifications, provide additional oversight and save money by negotiating favorable terms. Re-negotiation, particularly in adversarial circumstances, requires a strong understanding of available leverage. Sometimes, that leverage is chiefly financial. Often, it can also include components of law, technology and public opinion. A complicated negotiation, like restructuring a consortium's basic service agreement with a facility owner and/or operator, would benefit from the services of professionals in several fields. There are circumstances where incentive based contracts for these services may be appropriate.

Regulatory Issues -- Negotiations to resolve compliance or other regulatory issues, arising from operations or changes in law, require a thorough understanding of the relevant agencies and issues. Since these matters often arise in a very public and, therefore, political climate, it is critical to identify management consultants with proven communication and dispute resolution skills. The cost of not mitigating these issues immediately and effectively can be significant.

Incentive based contracts are rarely appropriate in these circumstances. While there often is a financial component to these issues, they also introduce the notion of the public interest. The appearance of the consultant arguing against the public interest, for financial gain, is unseemly.

Procurement Services

Procuring ash management/recycling services, waste handling/processing equipment or exotic chemicals are typical responsibilities of a waste-to-energy consortium. It is critical to draft clear, focused RFPs/RFQs to maximize competition for the consortium's business. When possible, holding preproposal conferences, reviewing potential vendors' assessments of the current market and revising procurement documents accordingly, when appropriate, are valuable steps. It is important to be familiar with similar, recent procurements elsewhere, while tailoring a particular consortium's procurement scope to its unique characteristics. Management consultants, with strong technical and business expertise, can strengthen these efforts. There are some fairly clear instances, where incentive based contracts can be appropriate for these services, discussed below.

Marketing Services

With highly competitive waste markets in many areas, selling excess capacity is critical to the bottom line of many consortia. Unfilled capacity can mean increases in tip fees to offset revenue loss. Management consultants, with effective waste market identification and penetration strategies can help maintain maximum throughput.

This is a very natural area for incentive based contracts. Goals and current baselines can be readily established and are clear. Improvements are reasonably easy to measure.

INCENTIVE BASED CONTRACTS FOR MANAGEMENT SERVICES

As mentioned in the introduction, a great deal of money is at risk in each of the areas just described. In cases where an objective is clear and unequivocal, an incentive based contract can make a lot of sense. For example, a given consortium's facility currently recovers 50% of the ferrous and receives a contractually established rate for the metal. An incentive based contract could be entered which would pay a management consultant an escalating amount for net financial improvement. If the management

consultant increases the metal recovery rate and/or the rate received for the material to an agreed benchmark, after cost, then a known return will accrue.

In the current example, there are many initial parameters and potential variables that must be pinned down. How much metal is in the stream currently? Will that amount change because of recycling or other external parameters? What actual percentage is currently being recovered? Once they have been established, with provision for externally imposed variables, the project goal can be fairly clearly defined. More importantly, the benefit to all consortium members is clear and mutual.

Generally, procuring well-delineated goods and services can also be appropriate for incentivized arrangements. Management consultants will be motivated to generate the broadest competitive interest and negotiate the most favorable contracts. As long as the client's interest is well known, adequately defined and quantified, these contracts can produce good results.

It is always important, however, to assure that there is no ethical conflict for the management consultant. For example, scenarios where consultants could establish inadequate specifications for the sought goods or services, to keep cost low and maximize their incentives, must be avoided. In cases where there is any question as to the availability of generally accepted standards, independent review of the process should be a component of any incentivized contractual arrangement.

Problem Areas For Incentive Based Contracts

Finally, there are circumstances in which incentivized contracts should generally be avoided altogether. As discussed above, regulatory negotiations present special problems. More generally, however, problems arise when the goal, which defines the client's interest, cannot be clearly and adequately defined.

In a complex negotiation to restructure a consortium's service agreement, for example, it may be difficult to determine, in advance, what will be in the best interest of all the municipalities impacted. The governing rules of the consortium become critically important in cases where those interests diverge as the process develops. It may seem, at the beginning of a large project, that all member communities share the same final goal. However, as time passes, conditions can change dramatically, and all communities may not believe the direction of the project is in their best interest.

Important questions of fairness and equity can arise. For example, should a community, in the minority, continue to be responsible for paying incentives to a management consultant, serving the majority's interest against their own? What mechanism exists for a minority community to escape such expenditure of its funds?

If a consortium decides to enter an incentive based contract, in such complex circumstances, anyway, there are some steps that can be taken to minimize, if not remove, problems of the sort described above:

- Require unanimous votes on matters that can encumber incentive payments to the management consultant
- Establish, in advance, a mechanism allowing dissenting communities to opt out of a process, not in their best interest.

• Spend the time and energy necessary to discuss and define any complex consortium goals as explicitly and clearly as possible.

If it turns out these discussions are troubled and don't result in well defined goals, it is a pretty good sign that an incentive based contract for services would not be advisable. It would make more sense to contract for consultant services, at ordinary rates, to help clarify and define those objectives. If clear, unanimous goals emerge from that process and sufficient safeguards are established to protect minority interests, then incentive based contracts might be considered.

CONCLUSIONS

There are many very good reasons for regional consortia managers to seek specialized assistance with the many challenges they face. In some, well-defined projects, incentive based contracts with service providers can promote excellent results for the consortium. However, when the project is very large and complex, with goals that aren't sufficiently clear, regional consortia must be very careful before executing such contracts. At a minimum, contractual safeguards and remedies must be implemented. They must guard against any member ever having to pay incentives to a management consultant, to work against their own interest. If such safeguards are not possible or practicable, the incentive based contract should be avoided altogether.

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The Waste-to-Energy Sector and the Mitigation of Greenhouse Gas Emissions

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INTRODUCTION

The waste-to-energy sector provides one important avenue for the United States to reduce greenhouse gas (GHG) emissions. The purpose of this paper is to highlight the significant GHG reductions capable of being achieved by the waste-to-energy (WTE) sector through avoided fossil generation and reduced municipal landfills. The paper begins with a review of the current voluntary reporting mechanism for "registering" GHG reduction credits under section 1605(b) of the Energy Policy Act of 1992. The paper then provides an overview of possible emerging international and domestic trends that could ultimately lead to mandatory targets and timetables for GHG mitigation in the United States and other countries. The paper ends with an analysis of the GHG benefits achievable by the WTE sector, based on the section 1605(b) report filed by the Integrated Waste Services Association (IWSA) on the GHG emissions avoided for year 1995.

MECHANISM FOR REPORTING GREENHOUSE GAS EMISSIONS REDUCTIONS

On October 19, 1994, the Department of Energy (DOE) released final guidelines for reporting GHG emissions as required by section 1605(b) of the Energy Policy Act of 1992. These guidelines include six technical support documents for accounting and reporting GHG reductions from each of the following sectors: electricity supply, residential and commercial buildings, transportation, forestry, and agriculture. Entities may wish to report GHG emissions in order to gain recognition, to establish a public record for emissions and reductions for future reference, and to demonstrate support for voluntary approaches to achieving environmental policy goals. Participation in the voluntary reporting program could help to document the significant contribution that the waste-to-energy sector makes to meeting the United States' commitment to reducing GHG emissions.

Although DOE has created an extremely flexible program with many options for the voluntary reporting of GHG emissions and reductions, the agency also encourages the submission of as comprehensive a report as possible. A comprehensive report would include information about: the entire organization and all its GHG activities, historic baseline emissions data for 1987 through 1990, annual emissions, emissions reductions and carbon sequestration projects, other participants in the project, emissions factors used to determine reductions, data on secondary and indirect impacts of the mitigation project, assumptions about the project, and data sources.

DOE pressure for comprehensive reporting poses some risks for section 1605(b) participants. The failure to submit a comprehensive report could undermine the creditability of the underlying mitigation activity. Specifically, it is possible that participants will not receive the full credit and recognition for GHG reductions achieved if they submit only the minimum data required under the section 1605(b) program.

EPAct Section 1605(b) Reporting

Overview of Guidelines.

The guidelines are mandated under section 1605(b) of the Energy Policy Act of 1992, which requires DOE to provide guidance for the voluntary reporting of GHG emissions, their reduction, and carbon fixation achieved through any measure. The guidelines provide information on who may report, what

information may be reported, and how projects should be analyzed for reporting. As a general matter, the guidelines are extremely flexible and are structured in such a way as to promote broad participation. This flexibility should enable utilities to receive credit for GHG reductions achieved through a wide range of measures for mitigating GHG emissions.

Forms for reporting have been designed to suit the guidelines' flexibility. Form EIA-1605 allows individuals to give a detailed account of GHG emissions, reductions, and carbon sequestrations for the entire entity (e.g., corporation, facility) or for specific GHG mitigation projects (e.g., GHG reductions achieved through supply-side efficiency improvements). To accommodate those entities wishing to minimize the reporting burdens, DOE has developed Form EIA-1605EZ, a simplified form that only requires a brief synopsis of the GHG emissions reductions achieved by a specific project in the most recent calendar year. Completed forms are submitted to DOE's Energy Information Agency (EIA), which is charged with the responsibility of administrating the section 1605(b) reporting program and maintaining the GHG registry (e.g., database).

As indicated above, the guidelines are separated into eight sections: a general guidelines section and six technical support documents dedicated to issues particular to a specific sector or activity area. The discussion below provides a brief summary analysis of the general guidelines and the technical support documents related to the electric supply sector.

Method of Reporting.

Who May Report. The guidelines provide that the following groups or individuals may report: any U.S. citizen or resident alien; any company, organization, or incorporated group; and any federal, state, or local governmental entity. These entities may report their reductions or sequestration efforts as long as they can "define a project and report physical data in enough detail to quantify results of the activity." No minimum reporting threshold is established in order to encourage broad participation and participation by small-scale initiatives.

The DOE rule permits the reporting of activities undertaken in association with others, such as efforts by a utility to implement demand-side efficiency improvements at industrial or commercial facilities (e.g., installation of high-efficiency electric motors or green lighting). In order to avoid double counting, reporters must identify other parties participating in the activity. The rule also allows the reporting by a third party or trade association. As will be discussed below, IWSA has submitted a section 1605(b) report for the entire WTE sector.

What Information May Be Reported. Gases covered in the DOE reporting program include carbon dioxide (CO₂), nitrous oxide, methane, and halogenated carbon substances (e.g., CFCs, HCFCs, PFCs). DOE will count both direct and indirect emissions reduction and sequestration activities related to these four gases. DOE decided against limiting the program solely to direct emissions (which arguably would be more manageable and transparent) in favor of the flexible approach reflected in the guidelines. Direct emissions are those which result from fuel combustion or other processes that release greenhouse gases on-site. Indirect emissions are those which occur off-site from the party whose activities caused the emissions to be generated elsewhere. For example, a manufacturer would report as indirect the emissions associated with the electricity purchased from a utility used to light its assembly plant, since

the production of that electricity had caused emissions elsewhere. In the case of the WTE industry, the principal indirect GHG effect is the avoidance of CO₂ and methane from landfills.

International activities are included in the types of activities that may be reported. The same criteria will be applied to international activities as domestic activities. Difficulties may arise in analyzing the international activity, such as determining the reference case or obtaining credible data, so special attention should be given to direct and indirect effects of these activities.

How to analyze projects for reporting. The project may consist of all emission-producing activities of an organization, several activities, or only one activity. Every report must: (1) define the reference case to use as a basis for comparison with the project, utilizing historic data and/or use projected or modeled data; (2) identify the project's major or minor primary and secondary effects; (3) choose an estimation method using default data or measured/estimated data for the reference case and the project; and (4) report reductions. The general guidelines illustrate the project analysis process in three case studies: an industrial cogeneration project, an energy-efficient project in a large office building complex, and the purchase of new solar-powered electricity generating equipment. These case studies illustrate the varying levels of detail and analysis that can be included in the reports.

Baseline or Reference Case. In order to define the reference case (e.g., the emissions level against which to measure the effects of the project), DOE suggests using either historical data or projected data. Historical data would involve averaging emissions for some previous year(s). An average could be taken from the 1987 to 1990 period, or from some year(s) more accurately reflecting normal operations. Projected data would involve extrapolating the reference case from past trends and external data to determine what emissions would have been in the year in which the project's effects are being measured. This process may involve modeling and adjusting for growth over time. DOE also suggests estimating the emissions per unit of production using historic or current-year data and adjusting for future growth by multiplying this emission rate by the rate of production in the reporting year.

Estimation Methods. DOE provides guidance for choosing the method for estimating emissions levels for both the reference case and the project case to determine emissions reductions. The guidelines and supporting documents recognize three categories of data: physical data, default data, and reporter-generated data. Physical data describes the activities involved in the project. Default data includes emissions factors and stipulated factors. Stipulated factors allow the conversion of physical data about the project into estimates of changes in energy use, GHG emissions, or carbon sequestration. Emissions factors are provided in the specific sector supporting documents, described below.

Projected Mitigation Future Years. In certain cases, the mitigation activity may result in greater GHG reductions in future years than in the current reporting year. One obvious example is a forestry project that will sequester carbon dioxide over a thirty-year period. Although some carbon sequestration may be achieved during the initial year, most of the sequestration will accrue over the life of the forestry project. This is also the case for most WTE projects. As explained in the last section of this paper, the combustion of municipal solid waste provides the greatest GHG benefit by avoiding landfill methane and CO₂ over a 20-to-30 year period. In the final guidance, DOE recognized the problems that could ensue from a purely retrospective GHG reporting system (e.g., reporting only GHG emissions actually

reduced, avoided, or sequestered during the current reporting year). As a result, the final guidelines allow for the reporting of the GHG emissions mitigated for future years as well as the current reporting year.

Electricity Supply Sector. Utilities have the flexibility of reporting for their entire system, one specific project, or at some level in between. The "supply sector" technical document is intended to assist utilities in determining an appropriate scope for the reporting of their projects.

The DOE identifies the following types of emissions reduction measures that will be creditable under the section 1605(b) program:

- fuel substitution;
- direct carbon removal by "cleaning" the fuel prior to combustion or by "scrubbing" the emission stream following combustion;
- energy efficiency improvements in electricity generation, conversion, and transfer; and
- energy efficiency improvements in the end-use equipment (<u>e.g.</u>, reducing energy demand).

The guidelines presents a standard methodology for estimating reductions from such projects, activities, and measures. The method is applicable to both carbon dioxide and non-carbon dioxide greenhouse gases and can be used to compute emissions from carbon content or from various technologies employed in the electricity supply sector.

FUTURE TRENDS

International

The Framework Convention on Climate Change is an international agreement that was signed by the U.S. and more than 150 nations at the Earth Summit in Rio de Janeiro in June 1992. The agreement committed signatory nations to take non-binding actions to reduce GHG emissions to 1990 levels by 2000. Emission levels for post-2000 were to be determined through subsequent negotiations following the Rio Convention.

The first of these negotiations -- the First Conference of the Parties of the Framework Convention on Climate Change -- occurred in Berlin in April 1995. At this conference, the parties agreed to launch a two year process to define actions and advance commitments to reduce GHG emissions in the post-2000 time frame. This agreement later became known as the "Berlin Mandate." The Second Conference of the Parties was held in Geneva from July 8-19, 1996. The Third Conference of the Parties is scheduled for December 1997 in Kyoto, Japan.

Geneva Conference. At the Geneva Conference, U.S. officials announced that for the first time the U.S. will seek to set verifiable and binding targets to reduce GHG emissions through an international agreement. On July 17, in a speech at the Second Conference of the Parties of the Framework

Convention on Climate Change in Geneva, Tim Wirth, Under Secretary of State for Global Affairs, stated that the U.S. is committed to reducing greenhouse gases, and that only through the setting of binding targets can progress toward emissions reductions be assured. Further, the setting of targets to reduce greenhouse gases must be achieved through maximum flexibility of implementation, which may include joint implementation and trading mechanisms. However, the U.S. proposal does not outline specific targets or goals for emission reductions -- specific numbers are to be decided through international negotiations at a later date.

In his speech, Wirth outlined the goals of the U.S. policy in seeking a binding agreement:

- the U.S. will focus on outcomes that are "real and achievable" and that will seek measured adjustments so that all nations may reduce emissions in an "economically sensible manner";
- the U.S. will seek market-based solutions that are flexible and fair and will not accept proposals that seek to gain an economic competitive advantage;
- the U.S. will seek a proposal that will require the participation of all nations, developing and developed, to take steps to limit emissions (the U.S. will continue to provide technical assistance to nations to help them reduce greenhouse emissions); and,
- the U.S. will seek to set verifiable and binding medium-term targets to reduce greenhouse gases.

In addition to outlining the guidelines for the U.S. proposal, Wirth criticized other proposals that have been submitted as being neither "realistic nor achievable." Further, Wirth underscored the importance of flexibility in the implementation of global emission reductions and that governments should be able to decide how they will achieve the reductions.

International Panel on Climate Change Report. Also presented in Geneva was a report by the Intergovernmental Panel on Climate Change (IPCC) on the environmental and socioeconomic impacts of climate change. Released in 1995, the IPCC report concluded that human activities have an adverse affect on climate change. These findings were officially accepted and supported by the Clinton Administration during the Geneva Conference.

Ministerial Declaration and Future Climate Change Activities. On the last day of the Geneva Convention, a ministerial declaration was adopted that calls for, or encourages, the development and establishment of legally binding post-2000 limits on GHG emissions. Provisions within this agreement, the "Geneva Declaration," encourage the development of legally binding emissions targets for developed countries and emissions reduction commitments from developing countries; endorse the conclusions of the IPCC report; support a U.S. recommendation that legally binding medium-term emission targets be pursued; and, urge expedited development, application, diffusion and transfer of climate friendly technology.

In December 1996, the Ad Hoc Group on the Berlin Mandate (AGBM) met to begin negotiations for emission reduction targets suggested in the Declaration. At that meeting, Parties to the Convention were asked to submit draft proposals for the climate change agreement for review at the next round of negotiations.

Move Towards Post 2000 Targets. The next negotiation session on the U.N. Framework on Climate Change was again held under the auspices of the AGBM in Bonn, Germany from February 25 to March 7. At that time, all draft proposals for a protocol, or other legally binding instrument, were reviewed. The 150 nations participating in the meeting failed to outline the bases for a Protocol to the U.N. Framework on Climate Change. There was no agreement on a timetable for reduction of GHG emissions, a CO₂ reduction target, or on whether reductions should be achieved through coordinated international policies or through policies determined by the individual countries. In the next step, the proposals will be combined into one document which will be distributed by June 1 and reviewed at the next AGBM meeting, scheduled to be held on July 28 - August 7 in Bonn, Germany.

Several different proposals were submitted for treaty negotiations. In January, the United States unveiled its proposal for tradeable emissions budgets to reduce the level of GHG emissions (see discussion below). In a move which surprised observers, the European Union (EU) agreed to call on industrial nations to cut GHG emissions by 15% by 2010. However, member countries were able to agree on country-by-country targets for only a 10% reduction. The remaining 5% reduction would be negotiated later. Other countries, notably the Pacific island states under threat from a rising sea level caused by global warming, are demanding a 20% reduction by 2005.

All of these proposals were criticized. The United States' proposal to create a system of tradeable GHG emission credits received a cool response from delegates and was attacked by environmental groups as "unintelligible, unpopular and extremely complicated." The United States and Japan claimed that the EU's proposal was not a firm commitment, but was simply an international negotiating position. In addition, the proposal was criticized because the proposed 15% reduction would be met by the EU as a whole, allowing some member countries to actually increase their emission levels. Industrialized nations reject the possibility of a 20% reduction on GHG emissions. Some industrialized nations continue to oppose the imposition of binding targets only on developed nations, claiming such a regime would allow developing countries an unfair trade imbalance because of the additional cost of goods produced under an emissions cap.

After the Bonn meeting, the United States proposed that Japan and the United States form a bilateral joint initiative to lead the world community in solving the global warming problem. Specific details of such a proposal have not been released, but Vice President Al Gore has stated that the proposal could be the basis for climate change negotiations later this year.

One final negotiating session is scheduled for October 20-21 before the Third Conference of the Parties to the UN Framework Convention on Climate Change meeting scheduled for December 1-12, 1997 in Kyoto, Japan. At the Third Conference of the Parties, the Parties will decide on what provisions will be included in a final agreement on climate change. The United Nations has insisted that the final agreement must include binding commitments to limit greenhouse gases.

National

U.S. Draft Climate Change Proposal. On January 17, the U.S. submitted its draft protocol to the United Nations for consideration in the climate change negotiations. The U.S. draft document proposes a number of provisions that the U.S. would like included in the final climate change agreement, including the strengthening of commitments by developing countries and the creation of emission trading provisions for developed countries.

The U.S. draft proposal includes a "trigger mechanism" that calls for the creation of a negotiation process that would include binding provisions for developing countries to reduce emissions to be completed by the year 2005. This negotiation process would take place through a separate round of negotiations after the Third Conference of the Parties is complete. The current climate change proposal under consideration by the Parties requires developed nations to take mandatory steps to reduce emissions, while developing countries are encouraged on a voluntary basis.

Further, the U.S. draft proposal would require developing countries to undertake the following:

- prepare annual reports on emissions inventories and report on steps to reduce emissions;
 and,
- identify and implement measures, not yet specified, to mitigate emissions.

For developed nations, the draft protocol proposes:

- to establish an "emission budget" for each country. The emission budget would encompass a multiple year period (or "budget period") in which a country would be allowed to "bank" emissions credits not used during the current period for future use, to "borrow" emissions credits (with a penalty) from a subsequent budget period, or trade emissions. The draft does not specify the size of the emission budgets (i.e. emissions allowances) or the duration of the budget periods (yet to be determined). The U.S. is currently considering budget periods between 3-10 years. In addition, the U.S. draft proposal states that after the first budget period and corresponding emission budget allowance, subsequent emission budgets must be less than or equal to the previous emission budget;
- to establish a process for reviewing developing country reports and improve emission reduction strategies;
- to allow emissions trading between countries with emissions budgets;
- to permit joint implementation between developed and developing countries, through which developed countries would receive emission credits for transferring energy efficient technology to developing countries; and,
- to create a national system for measuring emissions and reporting annually on measurement, compliance and enforcement practices for the relevant budget period.

Current political climate. In response to the U.S. policy shift toward verifiable and binding targets to reduce GHG emissions, both the Senate and House Commerce Committees held hearings in September on the status of the climate change negotiations.

At the hearings, House and Senate Members expressed concern regarding the Administration's position in the climate change negotiations. Several Members criticized the Administration for acting hastily on this issue without sufficient scientific data and for supporting positions that could harm the American economy and cost American jobs. Further, Members of Congress disagreed with any policy that would set binding emissions reduction targets for developed, but not for developing, countries.

The response to the new draft protocol has been lukewarm in the United States. Environmentalists feel that the unanticipated move toward flexibility in reaching emissions targets is a step backward. They also claim that the logistics of implementing an emissions trading plan, and then verifying its success or failure, will prove to be difficult. Industry, on the other hand, urges the administration to analyze the economic impacts of current climate change policy before committing itself to targets and time tables. However, in February more than 2,000 economists, including six Nobel Laureates, signed a statement declaring that preventive, market-based steps are necessary in light of the significant environmental, economic, social and geopolitical risks of global climate change. Such consensus may influence U.S. policy makers.

Committees in the House and Senate have stated that they intend to hold hearings on global climate change during the current Congressional session. Senator John Chafee, Chair of the Environment and Public Works Committee, is calling for nations to met flexible but binding emissions targets for greenhouse gases. While observers question whether a Republican-controlled Congress would ratify a treaty containing legally binding targets, the United States may be under increasing international pressure to make such a commitment in light of the ranking by the World Wildlife Fund of U.S. efforts to reduce CO₂ emissions as the absolute worst of the 20 major industrialized nations.

WASTE-TO-ENERGY IS CAPABLE OF ACHIEVING SUBSTANTIAL GREENHOUSE GAS EMISSIONS REDUCTIONS

There are approximately 135 municipal waste combustion (MWC) facilities currently operating in the United States. Most of the MWC facilities (91) are waste-to-energy plants that generate electricity or steam by feeding mixed municipal waste into large furnaces dedicated solely to burning trash. In a limited number of cases (23), the MWC facility generates electricity or steam by burning a refuse-derived fuel (RDF) that has removed recyclable and unburnable materials and then shredded or processed the remaining trash into a uniform fuel. In addition, there exist 11 RDF processing plants that provide RDF not only for furnaces dedicated to burn RDF, but also boilers that burn fossil fuel or other waste fuels. If

^{1/} In 21 cases, the MWC facility is an incinerator that combusts trash but does not recover energy from waste.

This section discusses several ways in which one might expect waste-to-energy facilities to mitigate GHG emissions. The first is the CO₂ that would have been emitted from the combustion of fossil fuel in order to generate electricity or steam. The second is the avoidance of methane and CO₂ that would have been emitted if the municipal solid waste had been disposed in a landfill instead of burned in MWC facility. As the discussion below indicates, the greatest opportunity for GHG mitigation exists from avoiding landfill gas emissions as well as ash reuse for various commercial purposes. The discussion below is based on IWSA's section 1605(b) report submitted to the EIA on GHG emissions avoided by the WTE industry for the year 1995.²

Avoided Fossil Generation.

One important benefit of waste-to-energy facilities is that they displace the use of fossil fuel for generating electrical power and steam. In 1995, for example, the waste-to-energy sector combusted over 31 million tons of municipal waste. The combustion of this fuel source resulted in the generation of 2650 megawatts of electricity every hour^{3/} and the export of nearly 1.4 million pounds of steam every hour. As a point of reference, this energy output is sufficient to meet the power needs of 1.2 million homes and businesses across the country. In addition, the combustion of this municipal waste avoided the use of about 30 million barrels of crude oil or almost 9 million tons of bituminous coal that would be necessary to generate an equivalent energy output.

The avoidance of fossil fuels in such a substantial amount has many clear energy, economic, and environmental benefits for the nation. Unfortunately, a <u>direct</u> net reduction in GHG emissions is not one of them. Using year 1995 as an example, waste-to-energy facilities emitted 31.3 million tons of CO₂ emissions to generate 2650 megawatts per hour. This represents a CO₂ emissions increase of slightly over 9 million tons, when compared against the CO₂ levels that would have been emitted from replacement coal-fired generation. In assessing the <u>total</u> GHG consequences of WTE facilities, the analysis does not end with a simple comparison of WTE and power plant emissions. As the next subsection indicates, WTE facilities also produce a number of <u>indirect</u> GHG benefits by reducing the amount of MSW deposited in landfills. These benefits more than offset any marginal increase in CO₂ emissions resulting from MSW combustion at WTE facilities.

Avoided Landfill Gases.

CO₂ and methane are two greenhouse gases emitted from landfills as the organic waste decomposes over a 20 to 30 year period. The decomposition begins with aerobic bacteria consuming oxygen while converting organic substances to CO₂, heat and water. This process continues until all available oxygen is depleted. After oxygen depletion, decomposition of organic material continues under anaerobic

The authors would like to recognize the efforts of Maria Zannes, President of IWSA, and Greg Gesell, American Ref-Fuel, in preparing the section 1605(b) report submitted on behalf of the WTE sector for year 1995.

Assuming parasitic usage of 20 percent, the net power exported into the grid is estimated to be approximately 2120 megawatts of electricity per hour.

The combustion of 31.3 million tons of waste by WTE facilities results in about 31.3 million tons in CO_2 emissions. In contrast, coal-fired generation of an equivalent energy output results in 21.1 million tons of CO. The reasons for this decrease of slightly over 9 million tons result from more a favorable heating value, heat rate, and CO_2 conversion factor for fossil fuel sources than waste-to-energy facilities.

conditions. In the absence of oxygen, anaerobic bacteria begin digesting the waste and producing methane. Over the life of the landfill, organic materials will be broken down into CO_2 and methane at ratio of approximately 45:55.

The combustion of MSW avoids the release of both CO₂ and methane from landfills. In the case of 33.3 million tons of MSW combusted in 1995, this translates into the avoidance of about 9 million tons of CO₂ over the life of the landfill. Similarly, landfill methane will be reduced by about 4 million tons over the same time frame. It should be noted that methane has substantially higher global warming potential than CO₂. According to latest estimates, methane has a "heat-trapping" capacity that is some 24.5 times greater than that of CO₂. Applying this factor to instant example, the avoidance of 4 million tons of methane is equivalent to a CO₂ emissions reduction of 98 million tons. Table 1 provides a summary of these GHG benefits. It should be noted that some landfills collect and combust a portion of the emitted methane. A few landfills use this methane for the generation of electricity. (See below.)

The Environmental Protection Agency (EPA) has promulgated New Source Performance Standards (NSPS) and Emissions Guidelines (EG) for landfills under Title I of the Clean Air Act. The requirements of the NSPS and EG are basically the same, with the main difference being the timing of implementation and the lead agency. Specifically, EPA administers the NSPS which takes place immediately for affected new landfills, while states implement the EG for affecting existing landfills once States have completed and received EPA approval of their implementation plans. As a general matter, the NSPS and EG standards require the collection and destruction of landfill gases at those facilities with a design capacity of 2.5 million metric tons and with annual non-methane organic compound emissions of at least 50 megagrams per year. Once fully implemented, affected landfills will consist of approximately two thirds of the nation's total landfill capacity. Although some landfills may install an energy recovery system, most facilities are expected to install a flare system that will burn off the collected methane. Although such flaring systems will reduce the amount of methane released into the atmosphere from regulated landfills, the methane combusted by flaring will result in a CO₂ emissions increase of 24.1 million tons. Table 2 provides a summary of these GHG benefits assuming all (e.g., affected and nonaffected) landfills comply with EPA's NSPS and EG requirements.

Ash Reuse.

In recent years, the use of MSW combustion ash has begun to increase for the WTE sector. Fifteen facilities currently are using a portion of their ash either as landfill cover, road building material or artificial reefs. Ash reuse is underway in projects from Florida to Maryland, Minnesota, Pennsylvania, Massachusetts, Tennessee and New York. There exist many additional safe applications of MSW combustion ash, which could provide a useful solid by-product and result in avoidance of GHG emissions. One example is use of WTE ash by-products in many cement and concrete applications as a substitute for portland cement and natural aggregates. The manufacture of portland cement and the mining of natural aggregates require considerable amounts of fossil energy and produce CO₂ emissions. Replacing portland cement or natural aggregates with WTE ash will offset the amount of fossil energy consumed and associated GHG emissions.

^{5/} See 61 Fed. Reg. 49,9905 (March 12, 1996).

Although not included in IWSA's section 1605(b) report, the use of MSW combustion ash represents another important GHG mitigation benefit resulting from WTE facilities. This mitigation opportunity, however, will only taken on greater importance as potential barriers to ash reuse are removed. 61

CONCLUSION

WTE facilities have the capability of achieving substantial GHG benefits for the environment. This becomes especially evident when avoided landfill gases are factored into the GHG mitigation analysis. Using 1995 as an example, WTE facilities are capable of achieving what is equivalent to a net CO₂ reduction of up to 98 million tons. If the amount of MSW combusted annually were to triple over the near term, the amount of CO₂-equivalent emissions avoided could increase up to almost 300 million tons per year. Such reductions would substantially contribute to the Clinton Administration's efforts to meet current and future GHG mitigation obligations being negotiated by international treaty.

Many of these barriers are related to safety concerns about the MSW combustion ash. These concerns, however, are largely unfounded. Specifically, the WTE sector has been testing for the safe management of MSW combustion ash for over a decade. These studies consistently show that leachate from ash landfills is similar to salty ocean water, with metals content at about the same level as the standards set for safe drinking water. MSW combustion ash tested under federal guidelines has consistently shown to be non-hazardous.

Table 1. NET GHG Benefit for 1995.

Greenhouse Gas	Emissions in Equivalent CO ₂			
Avoided Emissions Resulting from WTE Sector				
Fossil Generation CO ₂	22 million tons			
Landfill Methane	98 million tons			
Landfill CO ₂	9 million tons			
Total	129 million tons			
Emissions from WTE Sector				
Fossil Generation CO ₂	31 million tons			
Net CO ₂ Benefit	98 million tons			

Table 2. NET GHG Benefit for 1995 (assuming all landfills comply with EPA's NSPS and EG requirements).

Greenhouse Gas	Emissions in Equivalent CO ₂	
Avoided Emissions Resulting from WTE Sec	ctor	
Fossil Generation CO ₂	22 million tons	
Landfill CO ₂	24 million tons	
Total	46 million tons	
Emissions from WTE Sector		
Fossil Generation CO ₂	31 million tons	
Net CO ₂ Benefit	15 million tons	

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TECHNICAL SESSION III Fluidized Bed Combustion

Detection Of Lateral Non-Uniformities In Fluidized Bed Combustors

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ABSTRACT

This paper addresses the detection of lateral non-Uniformities in fluidized bed combustors by a novel probe technique not reported so far, by using the horizontal differential pressure fluctuations associated with bubble flow. The salient features of these fluctuations are highlighted utilizing the simulated records obtained by considering the Davidson's pressure field around a bubble. The simulation approach is validated by a comparison with the measured vertical and horizontal differential fluctuations in a freely bubbling fluidized bed.

INTRODUCTION

The concept of 'fluidization' finds wide application in combustors and incinerators, and ,from an environmental view point they find wide acceptance. At present they are being pursued vigorously to handle a wide array of fuels including municipal solid waste.

The onset of bubbling in a fluidized bed followed by continuous growth and subsequent eruption at the bed surface, greatly influences solid circulation patterns and gas phase mixing which inturn affects various in-bed processes like heat and mass transfer, erosion of in-bed surfaces, attrition and elutriation. In addition the mobility of a fluidized bed is also affected by distribution of bubbles across the bed cross-section, for the presence of any lateral non-uniformities in bubbling can severely affect the yield of a reactor or a combustor. These non-uniformities can arise on account of uneven distribution of the fluid or partial blockage of distributor plate including the disposition of various internals present in the combustor. It is always desirable to monitor the development of such non-uniformities in the course of operation of large units.

There are various techniques¹ that have been developed for the determination of local bubble characteristics but most of them are limited either to a small depth of observation or are impractical in large beds or hot and corrosive environments of industrial fluidized beds or combustors.² Of late, the interpretation of differential pressure fluctuations in terms of fluidization parameters is receiving considerable attention because of its potential application in demanding environments. Some of the advantageous features of this technique include the inherent robustness of such probes, their amenability for cooling in high temperature applications, the enormous amount of information contained in the response of such probes³, their relative insensitivity to mechanical damage or fouling, the possibility of locating them even outside the system.⁴

Any probe system meant to detect lateral non-uniformities must be able to reveal the presence of bubbles on both sides of its location using as minimum a number of sensors as possible. In this context the present study focusses on the development of a novel probe technique utilizing horizontal pressure differential record which is highly sensitive to the directionality of bubble approach (i.e) from left or right, and, which so far has not been reported. Specifically this paper addresses the prediction and simulation of horizontal differential pressure fluctuations under the freely bubbling conditions. The effect of growth of bubbles and the presence of other bubbles are considered along with the randomness of the bubble traversing path by considering the pressure field around a Davidson's bubble. The resulting trends are discussed in relation to those obtained experimentally from a freely bubbling 2-D bed.

SIMULATION OF HORIZONTAL DIFFERENTIAL PRESSURE FLUCTUATIONS

Figure 1 shows a schematic of a typical bubble flow sequence past the horizontal differential pressure probe and the resultant response. Since it is difficult to quantify the pressure distribution around a bubble with wake in terms of an usable model, circular bubbles are considered here. It will be shown later that even with this approach the predicted patterns resemble very closely with the measured records taken under freely bubbling conditions.

Considering the pressure field around a Davidson's bubble⁶ the excess pressure distribution relative to that in the dense phase remote from the bubble can be written as

$$\Delta P_{e} = \Delta P_{s}[r/S] \cos\Theta \qquad r \leq R \tag{1}$$

$$= \Delta P_s[R^3/Sr^2] \cos\Theta \qquad r > R \qquad (2)$$

where

$$\Delta P_s = (\rho_s - \rho_f) (1 - \epsilon_{mf}) g S$$

The normalized pressure gradient recorded by the horizontal differential pressure probe can be related as

$$[\Delta P_e / \Delta P_s] = [\Delta P_e / \Delta P_s]_{right} - [\Delta P_e / \Delta P_s]_{left}$$
(3)

(either of the two can be taken as reference) and the variations of this for a random bubble trajectory depicted in Fig.2 can be predicted in time co-ordinates by

$$Y = U_y t \tag{4}$$

$$U_{br} = 2/3 \sqrt{g R}$$
 (5)

$$Tan\beta = U_x/U_y \tag{6}$$

$$r = \sqrt{E^2 + Y^2} \tag{7}$$

and
$$E = e - Y Tan\beta$$
 (8)

The effect of bubble growth is simulated by assuming

$$D = D_i + Y^{1.64} (9)$$

to account for a high rate of growth to study the effect of rapid growth on the simulated differential pressure change patterns (This extreme limit corresponds to a growth in size of 60mm across a bed height of 180mm as found in actual measurements⁷). The presence of an another bubble on the pressure change recorded at the probe tip is considered by a summation of the pressure field due to each one when the other is absent and vice versa. This approach is found to be able to predict the pressure patterns satisfactorily under vigorously bubbling conditions as also verified by high speed photography. 8

The effect of bubble interactions on the velocity of individual bubbles is taken through Clift and Grace model⁹, where for bubbles in vertical alignment.

$$U_{1} = U_{b\infty 1} + U_{2}R_{2}^{3} / (x_{d} + R_{1})^{3}$$

$$U_{2} = U_{b\infty 2} + U_{1}R_{1}^{3} / (x_{d} - R_{2})^{3}$$

$$= U_{1} + U_{b\infty 2}$$

$$x_{d} < R_{1} + R_{2}$$
(11)

EXPERIMENTAL

To compare the simulated horizontal pressure differential fluctuations under freely bubbling conditions, measured records ⁸ obtained using wall pressure taps on a two-dimensional bed (0.4m x 0.028m) with sand in the size range of 360μm to 550μm are used here. The excess gas velocity range is from 0.03 to 0.23 m/s. Two pressure transducers (HBM PD 0.1, SCANIVALVE 5PSID) coupled to carrier frequency amplifiers (HBM KTS-5) and connected to a microprocessor based signal analyzer (SM 2100B IWATSU) complete the instrumentation setup. The size of the pressure taps is 0.8mm I.D and they are arranged to record simultaneously vertical and horizontal pressure gradients and the adequacy of the time constant verified by changing the data sampling time interval. The data collected correspond to a transducer interrogation frequency of 409 HZ.

RESULTS & DISCUSSION

Effect of bubble growth and obliquity

The effect of bubble growth on the resultant horizontal pressure fluctuation is presented in Fig.3. For a vertically rising bubble, as is the case here, there is a transition from negative to positive gradients followed in between by a dwell period during which the gradient is zero. This dwell period actually represents the time interval during which both the sensors of the probe are engulfed by the bubble. The sensitivity of the probe response to the directionality of the bubble approach (i.e.) from left or right is amply clear by the fact that in the second case the transition is from positive to negative gradient, during the period of bubble to probe interaction. If the bubble starts growing considerably during its ascent and past the probe, its net affect is to alter the time scales involved rather than the nature of fluctuation.

Fig.4(a). depicts the effect of bubble growth for an oblique bubble to probe hit wherein the pressure gradients are always negative for a bubble approaching from left. Again the effect of bubble growth is only to alter the time scales associated with the bubble residence at the probe. For a bubble approaching the probe from right side, the resultant pressure gradients turn out to be always positive, as indicated in Fig. 4(b) for the case with bubble growth. This in conjunction with Fig.3 brings out the extra - ordinary sensitivity of a horizontal pressure gradient record to the directionality and obliquity of the bubble traverse. The deviation from this trend observed for the one without growth

corresponds to a case where the bubble while crossing the probe obliquely does not completely engulf the two sensors of the probe. This can be easily understood by referring to Fig.2 where in

$$L1 = (e+S) \cos\beta > R$$
and
$$L2 = e \cos\beta < R$$
(12)

Only when both L1 and L2 are less than R, the gradients would be either entirely positive like in Fig.4(b) or entirely negative as in Fig.4(a). However the obliquity can be easily discerned from the fact that there is an additional incursion from negative to positive as denoted by the region 'abc' which however is not going to be there for a vertically rising bubble.

Effect of the presence of other bubbles

A comparison of the simulated vertical and horizontal pressure gradient fluctuations for successive bubble flow sequence past the probe is given in Fig .5. This simultaneous comparison of both vertical and horizontal gradients will be used to validate the soundness and utility of the simulation approach by a comparison with experimental records for the same probe configuration.

Figure 5 shows that for an oblique traversing path horizontal gradients are positive as expected and discussed earlier. The vertical pressure gradients are well known and discussed elsewhere. The important point to note here is that when the peak in vertical gradient is lower, the peak in horizontal gradient would be higher and vice versa, as can be seen at time intervals A-a and B-b. In the simulation the bubble interaction is suitably taken into account.

Comparison with experimental records

An experimental record of vertical and horizontal pressure differential changes simultaneously taken is presented in Fig. 6. along with the probe configuration. For all the three bubbles marked by vertical gradient pulses A,B and C it can be clearly observed that when vertical gradient peak is larger, the horizontal gradient peak is lower and vice versa. The fact that the horizontal differentials are always positive tends to indicate that it is for an oblique hit with the bubble completely engulfing both the sensors of the probe on its way past the probe. The bubble approach path which can be inferred is from left of the probe. For this path and probes, the direct similarity confirms the simulated patterns are the ones resembling closely with those to be expected in freely bubbling beds.

The effect of closely spaced bubbles on the simulated gradients in both directions is noted in Fig.7. It reveals that for close spacing, the gradients at the nose of the following (trailing) bubble will be higher. The flow path is such that the trailing bubble completely engulfs the probe while the leading bubble tends to avoid one sensor of the horizontal differential probe. The sort of deviation observed here is already discussed in the context of Fig 4(b).

Figure 8 depicts the experimental recordings for close spaced bubbles in a freely bubbling bed. A comparison with the vertical gradient pulse marked D brings in the strong similarity for bubbles in

the process of coalescence. The gradient at the nose of the trailing bubble is greater for both the gradients. The only difference between Fig.7 and Fig.8 is that the size of the leading bubble is smaller for simulated records whereas it is larger for experimental case. Similarly the pulses marked A,B,C and E are for bubbles at various stages of coalescence.

Sensitivity of horizontal differential probe response

Figure 9 tends show the various utility of horizontal differential trace in the sense that it contains much more information than that contained in a vertical gradient record. For the vertical probe(sensor spacing = 6mm as that for horizontal) the flow path corresponds to a bubble flowing obliquely but roughly midway through the two sensors. The resultant gradient is almost symmetric but the point to be noted here is that, it can't reveal whether the bubble approached from right or left since in both cases the pattern would be same. However, the horizontal gradient while revealing that the bubble completely intercepted the probe while rising obliquely(as the gradients are either completely

bubble completely intercepted the probe while rising obliquely(as the gradients are either completely negative or completely positive) in addition gives the information that it approached from right side of the probe. For a bubble flowing exactly midway through the two sensors of the probe the two peaks in the horizontal gradient pulse will be equal to each other.

It is this feature which facilitates the detection of lateral non-uniformities by requiring the presence of bubbles on both sides of the probe for uniformity in bubbling. The salient features of the horizontal pressure gradient fluctuations could be incorporated in a bubble detection and fluidization quality monitoring program for on line detection of lateral non-uniformities.

Unambiguous bubble detection

By a suitable positioning of the sensors of the probe and taking advantage of the sensitivity of the horizontal gradients to bubble flowpath, it is possible to detect the bubble rather unambiguously. For example, Fig.10 considers a probe system employed by Dent etal¹¹ in a Fludized bed combustor. For a vertically rising bubble midway through the two sensors, the horizontal differential is zero and this feature can be used directly measure the bubble diameter in conjunction with the vertical gradient information for this probe configuration. For a bubble flowing eccentrically but vertically past the probe, the horizontal differential resembles that of a point pressure fluctuation when it avoids one or both the sensors of the probe.

CONCLUSION

Lateral non-Uniformities in fluidized bed combustors can be detected with the help of horizontal differential pressure fluctuations with least number of sensors. A single horizontal differential probe can be used to judge whether or not bubbling is there on both sides of the probe by incorporating the unique sensitivity of this probe response to bubble traversing path in a detection program. Whether the bubble approached from left or right side of the probe can be easily detected since there is a complete transition from one type of pattern to other which are easily distinguishable and can be taken into account. In addition it is also possible to generate more information regarding bubble properties, which is rather authentic and unambiguous.

The technique can be applied in a commercial unit either by making the horizontal differential pressure probes as part of the bed internals or by locating them just on the walls of the combustor. Detection of the presence of lateral non -Uniformities will lead to corrective actions and hencewith a better degree of uniformity, the average bubble size can be reduced. This will help in a lower level of elutriation from the top of the bed surface.

REFERENCES

- 1. N.P.Cheremisinoff, "Review of experimental methods for studying the hydrodynamics of gas-solid fluidized beds", <u>Ind. Eng. Chem. Process. Des. Dev.</u>, 25:329(1986).
- 2. O.Sitnai, "Utilization of pressure gradient records in fluidized beds with internals for bubble parameter determination", Chem.Eng.Sci. 37:1059 (1982).
- 3. C.M.Atkinson and N.N.Clark, "Gas sampling from fluidized beds A novel probe system", <u>Powder Technol.</u>, 54:59(1988).
- 4. Ghadiri, M., B. Carter, R.R. Rathbone, R. Clift et al., "Diagnosis of gas flow patterns in fluidized beds", in proceedings of the 4th International Fluidized bed Conference, London, Institute of Energy, 1988, pp I/3/1-16.
- 5. A.Venkata Ramayya, S.P.Venkateshan and A.K.Kolar, "Bubble detection with horizontal pressure gradient measurements in gas-fluidized beds", <u>submitted for publication</u>.
- 6. J.F.Davidson, "Symposium on fluidization-Contribution to discussion", <u>Trans .Inst. Chem. Engrs</u> 39:230 (1961).
- 7. A.V.Ramayya, S.P.Venkateshan and A.K.Kolar, "Large particle fluidization studies with a differential pressure fluctuation record" in proceedings of the 12th International Conference on Fluidized Bed Combustion, vol.2,1993, p 877-883.
- 8. A. Venkata Ramayya, Ph.D Dissertation, Indian Institute of Technology, Madras, India, 1991.
- 9. R.Clift and J.R.Grace, <u>Fluidization</u>; J.F.Davidson, R.Clift and D.Harrison, Eds. Academic press, London, 1985, pp .73-132.
- 10. A. Venkata Ramayya, S.P. Venkateshan and A.K. Kolar, "Estimation of bubble parameters from differential pressure measurements in fluidized beds", Powder Technol., 87:113(1996).
- 11. D.Dent, R.D.LaNauze, T.Joyce, V.Fulford and P.Peeler, "Differential pressure measurements Their application to measurement of fluidized bed Combustion parameters", in proceedings of the 10th International Conference on Fluidized Bed Combustion, vol.1, 1989, p.451.

NOMENCLATURE

D_p	Particle diameter,m
Е	Eccentricity of bubble centre from the sensor,m
е	Eccentricity at the instant bubble nose crossing the probe,m
H_{s}	Static bed height,m
R	Bubble radius,m
r	Distance between bubble centre and the point of interest,m
S	Sensor spacing of the probe,m
t	Time,s
U	Superficial gas velocity,m/s
U_{br}	Bubble rise velocity ,m/s
U_{boc}	Bubble rise velocity in isolation,m/s
X_d	Vertical distance between two bubble centres,m
Y	Vertical distance between bubble centre and plane of the probe,m
Z	Vertical distance from distributor plate,m
β	Obliquity of bubble centre with probe, degree
Θ	Angle subtended by the line joining the bubble centre and the sensor at a particular instant, degree
€	Voidage
ρ	Density,Kg/m ³
Subsc	ripts
f	Gas
S	Solid Minimum fluidization
m _f 1	Leading bubble
2	Trailing bubble
X	X Component
Y	Y Component

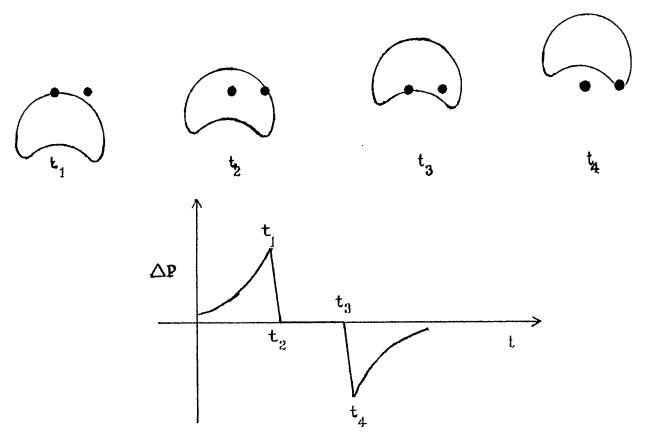


Fig: 1 A typical bubble to probe encounter and the resultant response.

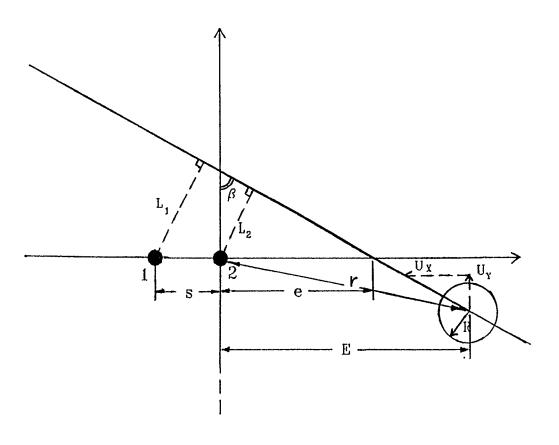
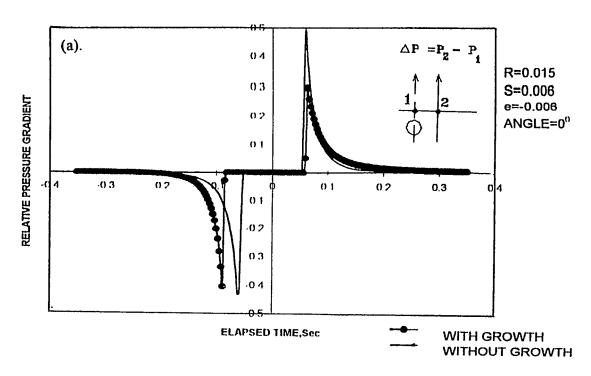


Fig.2 A typical random bubble trajectory.

SINGLE BUBBLE



SINGLE BUBBLE

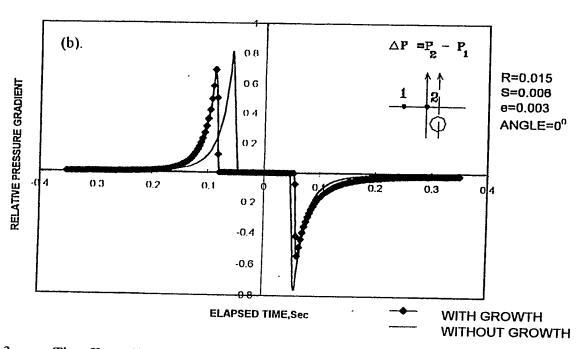
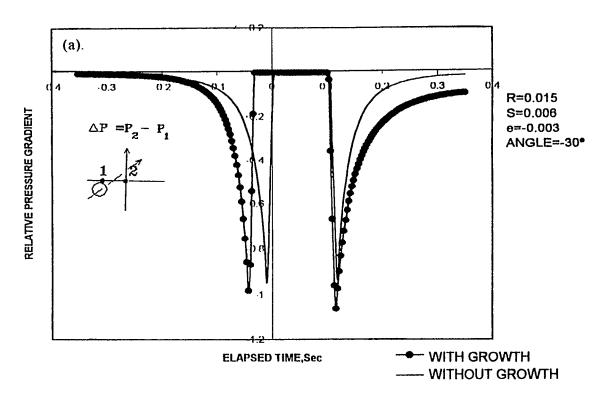


Fig. 3 : The effect of bubble growth on the horizontal pressure gradient record for a vertically rising bubble.

SINGLE BUBBLE



SINGLE BUBBLE

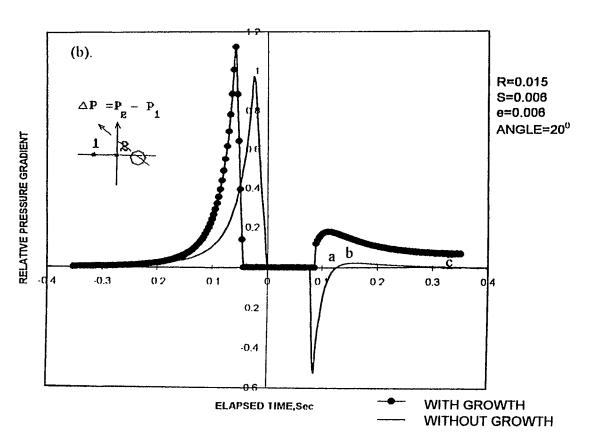


Fig 4 The effect of obliquity and bubble growth on horizontal pressure gradient fluctuation.



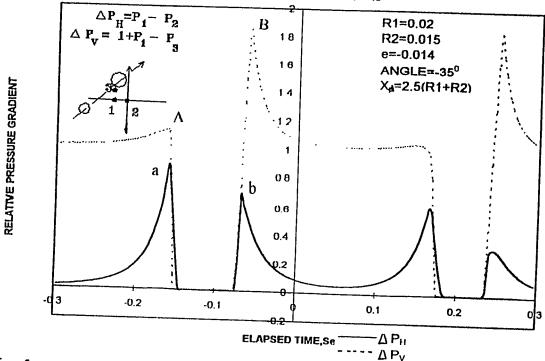


Fig. 5 Simulated vertical and horizontal pressure gradient fluctuations for wide spaced bubbles

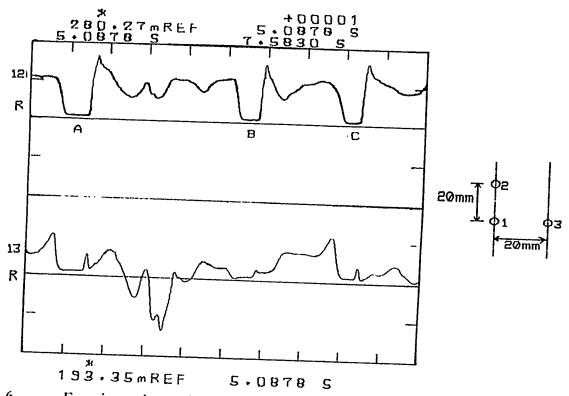


Fig. 6 Experimental recordings of the vertical and horizontal differential pressure changes in a freely bubbling 2-D bed (Sand, $D_p = 402~\mu m$, $H_s = 0.55~m$, Z = 0.32~m, U-U_{ntf} = 0.15 m/s) from 5.0878 s to 7.5830 s. At 5.0878 s, the vertical pressure differential is 280.27 mREF while that of horizontal pressure differential is 193.35 mREF.

TWO BUBBLES

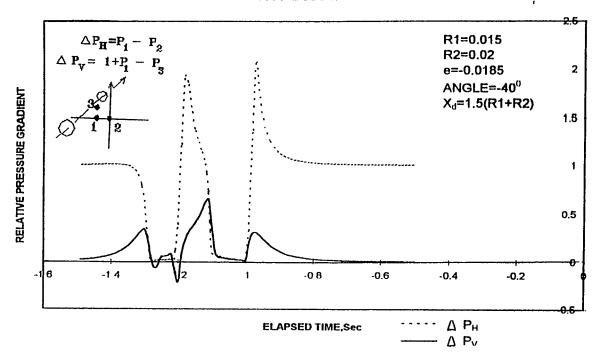


Fig. 7 Simulated vertical and horizontal pressure gradients for close spaced bubbles.

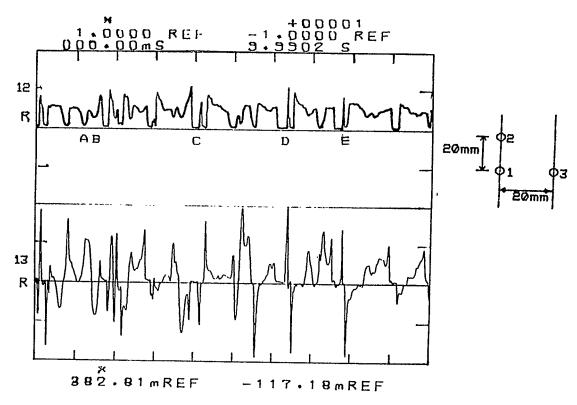
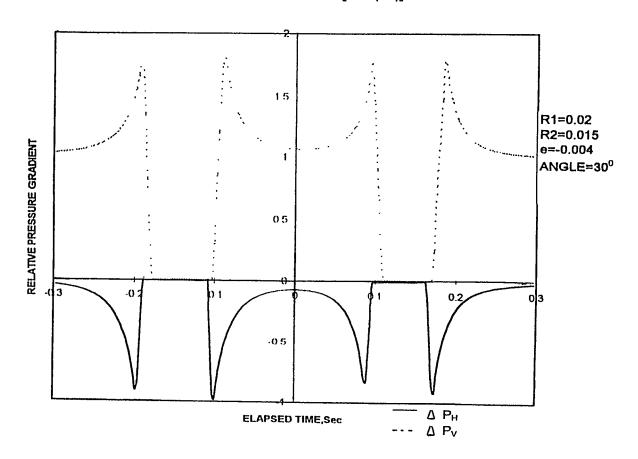


Fig. 8 Experimental recordings of vertical and horizontal differential pressures across a time interval of 9.9902 s in a freely bubbling 2-D bed (Sand, D_p = 402 μm , H_s = 0.55 m, Z = 0.32 m, and U - U_{mf} = 0.18 m/s). The scale is from 1 REF to -1 REF for vertical and between 382.81 mREF and -117.18 mREF for horizontal differential.

TWO BUBBLES [u2=f(u1)]



PATH OF THE BUBBLES

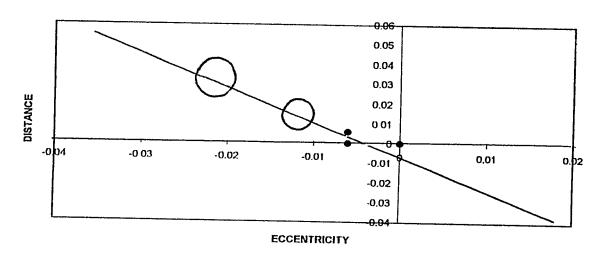
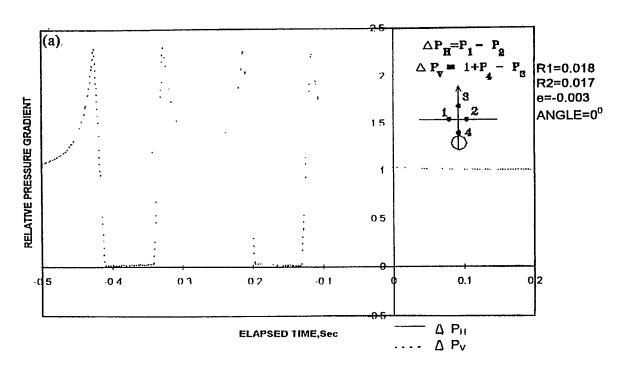


Fig. 9 . Simulated vertical and horizontal pressure gradient fluctuations for a bubble flowing obliquely and midway through the vertical differential probe.

TWO BUBBLES [u2=f(u1)]



TWO BUBBLES [u2=f(u1)]

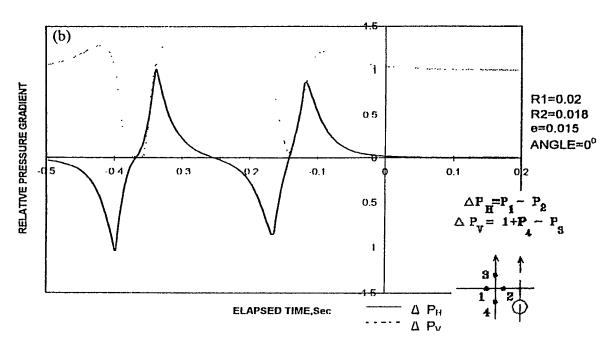


Fig. 10 Simulated vertical and horizontal pressure gradient fluctuations for (a) co-axial bubble hit and (b) eccentric hit.

Lime Enhances Moving Bed Filters for Mercury and Dioxin Control

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ABSTRACT

Moving bed carbon filters were developed and used in Europe to control mercury and dioxin emissions from municipal waste combustors (MWCs), crematories, hazardous waste incinerators and power plants. Although some operating problems such as a potential for fires to form in the carbon beds have caused operators some concern, more stable bed materials are presently being produced which can maintain the moving bed filter's capability in reducing mercury and dioxin emissions to very low levels while providing safe operations.

Märker Umwelttechnik GmbH (Märker) and Dravo Lime Company (Dravo) have developed a pelletized form of Sorbalit® that replaces the carbon in moving bed filters and is effective in removing mercury, dioxins, and acid gases without the inherent problems of using a carbon only adsorbent system. Sorbalit pellets are a formulation of portland cement, lime, carbon, and sulfur compounds. These pellets are currently being successfully applied in a hazardous waste incinerator, a foundry, a crematory, and a contaminated soil treatment plant.

This paper discusses the development of pellet technology and its application in moving bed filters. Safety aspects for the use of composite carbon/lime pellets will be discussed and a review of removal efficiencies among carbon alone, a composite carbon/lime material in a powder form, and composite carbon/lime adsorbents in a pellet form will be provided.

INTRODUCTION

The German air pollution regulations, like those in most of Europe, are intended to reduce pollution to the lowest technologically achievable value regardless of economic considerations. This contrasts sharply with U.S. EPA's approach which is to balance the economics of the waste combustor and health benefits of the country. The rationale for the stricter standards in Europe lies in the closer proximity of sources of pollution to residences and business centers, and accordingly requires them to be extremely good neighbors. It is not at all uncommon for local air pollution control regulators and owners of facilities to demand much lower emission limits than those required by the already strict German Federal regulations in order to facilitate community acceptance of the proposed project during permitting. This mind-set to reduce air emissions to the maximum has resulted in cases wherein air pollution control technology systems required as many as five separate control methods/elements to achieve very stringent and contractually required air emissions reductions. To illustrate, Table 2 presents a summary of the emission standards in the 17th BImSchV (present German Federal law) and compares these regulations with the U.S. EPA standards for MWCs with all values converted to metric units for ease of review.

As a positive consequence of this regulatory philosophy, major advancements to the technology of air pollution control were developed in Germany which have resulted in significant reductions in emissions of certain key pollutants such as dioxins, mercury, and acid gases. One innovative approach that was developed in response to the goal of requiring the lowest possible emissions was the use of a moving bed filter as part of a multi-component pollution control system. The moving bed filter, located in the final position, is referred to as a "police" or "polishing filter". The term "polishing filter" is used because the device completes the pollution control process by reducing already low levels of pollutants to extremely low values while catching any potential breakthroughs. Generally, such a device would not be practical, efficient

or economic in reducing high levels of pollutant emissions to code requirements. However, when used to reduce low levels of pollutant emissions, extraordinary results can be achieved. For example, say an upstream pollution control device such as a dry scrubber / fabric filter has reduced SO₂ emissions from 250 ppm to 10 ppm, then typically, the addition of a moving bed filter to the pollution control train downstream of the dry scrubber could reduce or polish the SO₂ emissions from 10 ppm to <1 ppm before discharging the flue gases to the atmosphere. Such results, although unimaginable only a few years ago, are indicative of the progress that can be made when the public demands and is willing to pay for state-of the-art technology when the possibility, however remote, of negative effects on human health are the alternative and money is viewed as an unlimited resource.

As illustrated in the above example, this innovative multi-component approach to air pollution control utilizing a moving bed filter technology has produced reductions in some pollutant levels on an order of magnitude lower than the already very low levels required by the regulations established in Germany by the 17th BImSchV, the Federal law in Germany which regulates the air emission standards for all waste management facilities.

The following is a summary of typical permit limits required by local government for various waste management projects implemented and built in Germany in the early 1990's. This summary illustrates the relentless ratcheting of German permit limits and contract specifications beyond those required by the 17th BImSchV.

Summary of Typical Permit Limits

Emissions @ 11% O2	17th BImSchV	Permit Limits	Contract Specifications
Dioxins ng/Nm ³	0.1		
Range	-	0.05 - 0.1	0.01 - 0.05
Typical	•	-	0.05
Mercury $\mu g/\mathrm{Nm}^3$	50		
Range	-	5 - 50	1 - 25
Typical	-	20	10
NOx mg/Nm ³	200		
Range	-	70 - 200	50 - 200
Typical	-	200	70 - 100

TECHNOLOGY OF MOVING BED FILTERS

Moving bed filters have been successfully applied to the control of both particulate and gas phase contaminants in flue gas streams from a host of industrial and municipal processes. Particulate matter is removed by impaction on the bed media which is generally spherical with diameters in the range of 2mm to 8mm. Gas phase components are removed by the selection of a chemically reactive filter medium appropriate to the gas phase component to be removed.

For example, carbon media has been proven effective in the control of heavy metals and organic emissions. In this configuration, moving bed filters have two primary elements, the filter media (activated carbon or lignite coke) to adsorb pollutants from the flue gas and a housing or moving bed filter to contain the carbon. In the moving bed filters, the carbon is constantly fed into the top of the housing structure and is removed from the bottom. Both cross-flow and counter-flow moving bed filter system configurations are available. In the cross-flow design, the contaminated flue gases flow across the vertically downward moving bed in a horizontal direction. In the counter-flow design, the contaminated flue gases flow in an upward direction through the vertically downward moving bed. Hybrid configurations which apply both cross-flow and counter-flow schemes have also been built. Accordingly, moving bed filters can be built in a variety of ways to be used as a single stage or as a multi-stage control system. These stages can consist either of a single vessel with multiple bed levels or of multiple vessels each dedicated to the control of a single contaminant or group of contaminants.

Figure 1 illustrates the above described configurations for moving bed filter systems for air pollution control. Typically in an initial stage, dioxin, mercury, HCl and SO₂ are removed. In a secondary stage and after ammonia (NH₃) is injected into the flue gases, NO_x can be removed by the carbon. As described previously, moving bed filters are often used after the primary acid gas (SO₂, HCl) control device such as a wet scrubber or spray dryer/fabric filter. This is to remove the majority of the acid gas (90 - 99%) using a calcium based reagent (\$80/ton) and thus minimize the usage of the very costly carbon (\$300 - \$900/ton) for acid gas control. Thus, as a practical matter and in terms of economics, moving bed filters are not used as a primary air pollution control system.

Moving bed filters offer the following advantages as pollution control devices:

- 1) Multiple pollutants can be removed in a single application.
- 2) They do not require a large pressure drop and can be retrofitted into many sites.
- 3) Carbon provides an excellent medium for the reduction of dioxins, organics, and mercury to well below permit limits.
- 4) In most cases, the used carbon can be injected back into the furnace and burned, thus eliminating a disposal problem and the associated disposal cost.

As pollution control devices, moving bed filters offer the following disadvantages:

- 1) The massive volume of carbon present in the filter is a potential fire hazard.
- Good SO₂ controls are required prior to the moving bed carbon filter. Since carbon has a high adsorption affinity for SO₂, this affinity coupled with the much higher amounts of SO₂ present in the gas stream from combustion sources will quickly use up the available carbon thereby letting mercury and dioxins pass through.

- 3) NO_x reduction capability is not as good as that of other control methods such as selective catalytic reduction technology (SCR). If high levels of NO_x control are required, (below 100 mg/Nm³) other more effective technologies such as SCR technology should be employed.
- 4) Handling granular carbon is messy and requires additional housekeeping. The use of composite carbon/lime pellets can almost completely mitigate this problem.

A SORBALIT CASE HISTORY

Märker was called by the owner of an industrial sludge incinerator and asked for help in solving several operating problems at the sludge plant connected with the plant's air pollution control system. The air pollution control system consisted of a dry scrubber with a fabric filter followed by a moving bed carbon filter. After careful review, the sludge at this facility proved to have a higher than normal sulfur and chlorine content which increased the acid gas quantities exiting the dry scrubber and entering the moving bed filter system. This resulted in a very high carbon usage rate since the moving bed filter system had been originally designed to control only mercury and dioxins. There were additional material handling problems at the plant resulting from the use of carbon.

To solve the problem, Märker developed a plan to replace the granular carbon (4mm) in the filter and replace it with a pelletized form of their Sorbalit technology. Sorbalit is a formulation of lime, activated carbon or lignite coke, and proprietary sulfur compounds. A binder such as portland cement or bentonite is used to strengthen the formulation in order to form pellets. The activated carbon component in the composite carbon/lime pellets ranges from 5% to 35% by weight. Photos of typical pellets are presented in Figure 2.

Composite carbon/lime pellets can be manufactured in a variety of sizes and shapes to meet the size of the filter media material handling requirements for the different manufactures of moving beds. The pellets in Figure 2 were produced by the "spinning disks" or "snow balling" technique. The nominal 4mm pellets on the left were made in Märker's laboratory in Germany using portland cement as a binder with composite carbon/lime. Dravo Lime made the 10mm pellets using bentonite as the binder. Both portland cement and bentonite have proven to be effective binders. They provide sufficient surface strength so that the pellets do not crush using screw feeder in the material handling process. The second property of the binder is that it is porus and permits flue gases to enter the pellet so that the carbon and lime can adsorb or absorb their respective pollutants.

The concept for the use of the composite carbon/lime pellets was that the lime would absorb the acid gases while the carbon adsorbed the dioxins and mercury. The only modification made to the moving bed filter was to replace the granular carbon with the composite carbon/lime pellets in the feed hopper. Test results of the air pollution control system demonstrated that the use of the composite carbon/lime pellets reduced the emissions to within specified levels on a sustained basis at expected composite carbon/lime usage rates. The composite carbon/lime formulation used for this application based on laboratory testing consisted of 10% lignite coke, 10% portland cement, and 0.25% sulfur compounds, and the balance hydrated lime.

Of prime interest to all parties during the development of the pellet technology was the confirmation that the binders would not affect the ability of adsorbents to control dioxin and mercury emissions. Unfortunately, Märker cannot presently test dioxin removal in the laboratory. However, Märker does have the capability to determine the effectiveness of different adsorbents with HgCl₂. Consequently in their

research laboratory, Märker compared formulations of their regular adsorbents containing 5% carbon and 95% hydrated lime with Dravo's pellet formulation which contained the same mixture plus a 5% bentonite binder. The test results show that there were no negative effects caused by the presence of the binder in the pellets on the Hg adsorption capacity of the composite carbon/lime. The test data are presented in Table 1.

These results are also encouraging as to dioxin adsorption in that Märker's past experience has been that Hg capture with carbon or carbon based sorbents is more difficult than dioxin capture under any given set of flue gas conditions. The literature is replete with many examples of field test data typically showing >95% capture rates of dioxins at concentration levels in the low ng/dscm range with concurrent and somewhat lower Hg capture rates of 85 - 95% when the Hg concentration levels were in the μ g/dscm range. As a result, testing of the sorbents for dioxin removal are expected to show higher levels of dioxin removal than Hg removal.

THE CARBON ADSORPTION PROCESS

The adsorption of mercury and organic materials such as dioxin into activated carbon and coke is controlled by the properties of both the carbon and the adsorbate, and by the conditions under which they are contacted. This phenomenon is generally believed to result from the diffusion of vapor molecules into the surface of the carbon. These molecules are retained at the surface in the liquid state because of intermolecular or Van der Waals forces.

As the temperature falls, or as the partial pressure of the vapor above the carbon rises, the average time that a molecule resides on the surface increases. So does the fraction of the available surface covered by the adsorbate. However, the carbon surface is not uniform and consists of sites whose activities vary. More active sites will become occupied first and, as the activity of the remaining available sites decreases, the adsorption energy will change.

The physical structure of activated carbon and coke is not known in detail, but it is believed to contain randomly distributed pores in the carbon, between which lies a complex network of irregular interconnected passages. Pores range in diameter down to a few angstroms, and provide a internal surface area from 300 to 1,000 m²/gram of carbon. The volume of pores at each diameter is an important variable that directly affects carbon performance.

Since adsorption takes place at the carbon-gas interface, the surface area of the carbon is one of the most important factors to consider. The second factor is the pore radius. However, the surface area must be available in the proper range of pore sizes. If too much of the area is available in pores smaller than 5 Å, many molecules will be unable to penetrate the pores and that area of the carbon will essentially be unavailable for adsorption. For most pollution-control applications, the surface areas of pores whose diameters range between 5 and 50 Å yield good efficiency rates because the relative pressure of the vapor is usually too low for the larger pores to become filled. At high relative pressures, however, the total pore volume becomes important because the macropores also become active.

The size of a molecule of mercury is approximately 3.6 Å and the dioxin molecule is 10 Å x 3 Å. Both molecules are adsorbed in different parts of the carbon particle. In theory, dioxins are collected in the

macropores (r > 25nm [250Å]) while the mercury is collected in the micropores (0.4nm [4 Å] < r < 1nm [10Å]). Dioxin, being larger, blocks the passages, preventing mercury from entering the micropores. To increase the mercury capture rate the amount of carbon used must be significantly increased, the surface area of the carbon must be increased, or sulfur added.

SULFUR'S ROLE IN MERCURY ADSORPTION

The addition of sulfur compounds to the process plays a major role in the adsorption of mercury but not in the adsorption of dioxin. Sulfur's role in the adsorption is two-fold:

- First, the sulfur compounds maintain the active state of the carbon. Activity is defined as the amount of open pores in the carbon. Sulfur's role is to keep these pores open and to allow the mercury to get into the sub-structure pores. The exact process in which the sulfur keeps the pores open has not been defined. One theory is that the sulfur reacts with water which is adsorbed or is on the surface of the carbon particles to form an acid that penetrates the pores.
- The second role for sulfur is to convert elemental mercury (Hg°) to a sulfate. Hg° is more difficult to capture than Hg₂Cl₂ (which is the predominant species in MWC emissions) or Hg₂SO₄.

Hg° accounts for 5 to 10% of the total mercury emissions from an MWC. For HWIs and fossil fuel fired power plants as well as other combustion sources however, elemental mercury can be 50% of the total mercury emission. Flue gas constituents such as SO₂ can further increase the elemental mercury content by reducing the dissolved HgCl₂ in wet scrubber liquors to Hg° which is consequently driven into the gas stream due to its poor solubility.

$$SO_2 + 2 HgCl_2 + H_2O \Rightarrow SO_3 + Hg_2Cl_2 + 2 HCl$$

 $Hg_2Cl_2 \Rightarrow HgCl_2 + Hg\dagger$

The adsorption capacity of carbon is affected by formation of sulfuric acid on the carbon owing to adsorption of the flue gas constituents SO₂ and H₂O:

$$SO_{2, gas} \Rightarrow SO_{2, ads}$$

 $SO_{2, ads} + \frac{1}{2}O_{2, ads} \Rightarrow SO_{3, ads}$
 $SO_{3, ads} + H_2O \Rightarrow H_2SO_{4, ads}$

Elemental mercury then reacts with the sulfuric acid to form mercurous sulfate (Hg₂SO₄) or in the presence of excess acid to form mercuric sulfate (HgSO₄):

$$\begin{split} 2 \; Hg + 2 \; H_2SO_{4, \, ads} &\Rightarrow Hg_2SO_{4, \, ads} + 2 \; H_2O + SO_2 \\ or \\ Hg_2SO_{4, \, ads} + 2 \; H_2SO_{4, \, ads} &\Rightarrow 2 \; HgSO_{4, \, ads} + 2 \; H_2O \; + SO_2 \end{split}$$

Since the lime component of composite carbon/lime removes the SO₂ from the flue gas, some adsorption capacity of the carbon for Hg° is diminished. The sulfur component in Sorbalit added during

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manufacturing, replaces the missing SO₂ and enhances the adsorption of Hg°. Mercuric chloride does not react with the sulfuric acid, but is dissolved in sulfuric acid.

FIELD TEST RESULTS

To date, composite carbon/lime pellets have replaced granular carbon in moving bed filters at a sludge incinerator (hazardous waste), a "green food" dryer, a foundry, and a soil treatment plant. Due to the competitive business these plants are in and the contractual obligations associated with the manufactures of the moving bed filters, Märker cannot presently publish the actual emission data. However, we can acknowledge that these facilities are currently meeting the strict German Federal emission regulations. For informational purposes Table 2 is a comparison in European units of Germany's emission regulation for waste management facilities and U.S. EPA's emission standards for new large MWC's. Table 3 is a tabulation of emission standard for hazardous waste facilities in Europe and Table 4 is a summary of U.S. EPA's proposed emission standards for hazardous waste facilities. Based on the test data currently available, the composite carbon/lime pellets used in a moving bed filter as a police filter will easily meet the U.S. EPA emissions regulations as well as the more stringent German and European standards.

The Institute of Gas Technology (IGT) has undertaken a development program of converting contaminated sludges from U.S. harbors and turn them into saleable "tiles". IGT has several combustion technologies that they employ to slag the harbor dredging and to destroy the organics which include PCB's and dioxins. In IGT's pilot plant test program, they wanted to ensure that they had effective methods to capture organics that were a result of incomplete combustion. IGT evaluated both a fixed carbon bed and a fixed bed of composite carbon/lime pellets to control the off gases from their process. At the time of the writing of this paper, the pilot plant test has been complete, however, the laboratory work still requires several months to confirm that European and U.S. experience coincide.

CONCLUSIONS

Carbon adsorption has been developed into a proven and effective method of controlling various hazardous pollutants to extremely low levels. The use of carbon as a collection medium in a moving bed filter has resulted in operating problems at various facilities. Typically for combustion sources, temperature excursions have resulted fires. Consequently, the selection and use of pelletized adsorbent formulations such as composite carbon/lime pellets has demonstrated the capability to replace activated carbon and to mitigate their safety and material handling problems although still in the developmental stage.

There are many other potential applications yet to be developed for composite carbon/lime's use as the adsorbent medium in a police or polishing filter as technology develops. At a U.S. EPA workshop on Dioxins held in Washington in December, 1996 a number of industrial and combustion sources of dioxin emissions were identified. Moving bed filters using carbon based sorbents could be potentially utilized widely in many of these industrial applications.

REFERENCES

- 1. W. Jockel, "Measurement and Monitoring of Mercury Emissions into the Atmosphere", German Environment Protection Journal, pp.46-51.
- 2. Official Journal of the European Communities, "Proposal for a Council Directive on the Incineration of Hazardous Waste", submitted by the Commission on March 23, 1992.
- 3. L. L. Sloss, "Emissions and Effects of Air Toxics from Coal Combustion: An Overview", Paper Presented at Managing Hazardous Air Pollutants: State of the Art, 13-15 July 1993, Washington D.C.
- 4. A. Licata, M. Babu, W. Carlson "The Application of Activated Carbon Enhanced Lime for Controlling Acid Gases, Mercury, and Dioxins form MWCs," American Chemical Society, Spring Meeting, New Orleans, 1996
- 5. J. Kilgroe, A. Licata, "Control of Air Pollution Emissions from Municipal Waste Combustors," National Waste Processing Conference ASME, Atlantic City 1996

Table 1. Sorbalit research test data.

	% Hg Removal	92.8%	97.2%	98.1%	%0.96	95.3%	%6′26	98.1%	97.1%
	Hg Total Quantity Out µg	0.49	0.19	0.13	0.27	0.32	0.14	0.13	0.20
	Hg Total Quantity In µg	6.8	8.9	8.9	8.9	8.9	8.9	8.9	6.8
u(Solution Metered g	4	4	4	4	4	4	4	4
HgCl ₂ Solution	HCI Content mg/g	125	125	125	125	125	125	125	125
H	Hg Content µg/g	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7
	Reactor Temp °C	180	180	180	180	180	180	180	180
	Adsorbent		Sorbalit with	5% lignite	COKE	Dravo Lime	pellets:	0.5% Sulfur	5% Bentoni te
Descrion	Conditions (Adsorbent Weight, mg)	250	250	250	Average Test 1	250	250	250	Average Test 2
	Test No.	1A	1B	1C	Aver	2A	2B	3C	Aver

Table 2. German and U.S. EPA standards for MWCs $^{(1)}$ (metric units corrected to 11% O_2).

Pollutant		Bundesgesezt	Germany Bundesgeseztblatt - 1990 All Sized Units		
SO ₂	mg/Nm³ % reduction Avg. time-hrs.	200 N/A 0.5	50 N/A 24	New Large Units 61 or 80% 24	
HCl	mg/Nm³ % reduction Avg. time-hrs.	60 N/A 0.5	10 N/A 24	28 or 95% 24	
PM	mg/Nm³ Avg. time-hrs.	30 0.5	10 24	18.3	
Cd	mg/Nm³	Included in Class I Metals ⁽²⁾	0.05	0.0153	
Pb	mg/Nm³	Included in Class III Metals	0.5	0.153	
Hg	mg/Nm ³ % reduction Avg. time-hrs.	Included in Class II Metals -	0.05 N/A >60 min.	0.061 or 85%	
Dioxi	n Furan ⁽³⁾ ng/Nm ³ TEQ ng/Nm ³ mass Avg. time-hrs.	- - -	0.1 ⁽⁴⁾ N/A >500 min.	≈0.2 ⁽⁵⁾ 13	
NOx	mg/Nm ³ Avg. time-hrs.	400 0.5	200 24	219 24	
CO	mg/Nm³ Avg. time-hrs.	100 1	50 24	89 4	

⁽¹⁾ All emission data based on mass burn water wall technology

Class I Heavy Metals: Cd & Tl
Class II Heavy Metals: Hg

Class III Heavy Metals: As, Co, Cr, Cu, Mn, Ni, Pb, Sb, Sn, & V

U.S. EPA dioxin standards are on a mass basis. There is no direct correlation to TEQs and only an approximate conversion can be made with a \pm error.

Germany requires a minimum combustion zone temperature of 850°C for 2 seconds.

U.S. EPA requires control of temperature at APC system.

Table 3. HWI emission guidelines for some European countries (mg/Nm 3 dry at 11% $\mathrm{O_2}$) .

Basis	Federal Republic of Germany Basis 17th BImSchV 11/90		Netherlands BLA 1993	EC D	irective
Measurement	Corrected only when >11% O ₂ 24 hr. avg max.½ hr.		Corrected at All Times max. 1 hr. mean	max.½ hr.	24 hr. Max
HCl SO (SO ₂ + SO ₃) HF NOx (NO ₂) CO C (organic) Particulate	10 50 1 200 50 10	60 200 4 400 (hr.) 100 20 30	10 40 1 70 (Boiler) 50 10 (8 hr. avg.) 5	10 50 2 - - 10 10	5 25 1 - 50 5 5
Heavy Metals					
Class I	Cd + Tl \sum = 0.05 (>.5 hr.)		Hg = 0.05 (8 hr. avg.)	$Cd + Tl \sum = 0.05$ (>.5 hr.)	
Class II	Hg = 0.05 (>.5 hr.)		Cd = 0.05 (8 hr. avg.)	$Hg = 0.05 \ (>.5 \ hr.)$	
Class III	Sb,As,Pb,Co,Cr,Cu, Mn,V,Sn,Ni $\Sigma = 0.5$ (>.5 hr.)		Sb,As,Pb,Co,Cr, Cu,Mn,V,Sn,Ni, Te, Se $\sum = 1.0$ (8 hr. avg.)	Sb,As,Pb,Co,Cr,Cu, Mn,V,Sn,Ni \sum = 0.5 (>.5 hr.)	
PCDD/PCDF (ng/Nm³)	0.1 I-TEQ > 500 min.		0.1 I-TEQ (8 hr. avg.)	0.1 I-TEQ ⁽⁶⁾ (8 hr. avg.)	
Combustion 1200°C >2 sec. Temperature		850 °C at $>6\%O_2$ >2 sec.	850° C at $>6\%O_2>2$ sec.		

⁽⁶⁾ Guideline only.

Table 4. U.S. EPA Emission Limits for Hazardous Waste Combustors (Proposed 4/96)

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Control of the contro

				Emission Limit (1)	nit ⁽¹⁾			
Pollutant	Averaging Time ⁽⁵⁾	Units	Incinerator	erator	Cemer	Cement Kiln	Lightweight Aggregate Kiln	t Aggregate In
			Existing	New	Existing	New	Existing	New
$ ext{PM}^{(7)}$	CEMS - 2 hours	mg/dscm (gr/dscf)	69 (0.030)	69 (0.030)	69 (0.030)	69 (0.030)	69 (0:030)	69 (0.030)
Dioxins / Furans (2) (6)	stack test	ng/dscm	0.20	0.20	0.20	0.20	0.20	0.20
Hydrocarbons (7)	CEMS - hourly	ppmv	12	12	20 (3)	20 (3)	14	14
ω 00	CEMS - hourly	ymdd	100	100	(4)	(4)	100	100
$HCI + CI_2^{(0)}$	CEMS - hourly	hmdd	280	<i>L</i> 9	630	<i>L</i> 9	450	62
Hg ⁽⁷⁾	CEMS - 10 hours	μ g/dscm	50	50	50	50	72	72
	stack test or	μ g/dscm	ı	1	1	55	12	5.2
Semivolatile Metals (Pb, Cd) (sum) ⁽⁷⁾	CEMS - averaging times vary	μg/dscm -	270 12 hours	62 10 hours	57 10 hours	60 10 hours	60 10 hours	60 10 hours
Low Volatility Metals	stack test or	μ g/dscm	ı	09	•	44	ı	55
(As, Be, Cr, Sb) (sum) ⁽⁷⁾	CEMS - 10 hours	mg/dscm	210	80	130	80	340	80
7000 Last assistant to 70% of 1000 Last and 1000 Last assistant to 1	C 000 F 20 200 100 100							

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All limits are corrected to 7% oxygen and 20°C.

TEQ basis.

The limit given is for the main stack, with the option of meeting a 6.7 ppmv by-pass duct limit.

The limit for the by-pass duct is 100 ppmv; there is no applicable CO limit for cement kiln main stacks.

Average time based on "rolling averages."

Based on confirmatory test under normal conditions.

Determined during comprehensive test (an operating condition that will result in higher than normal emissions).

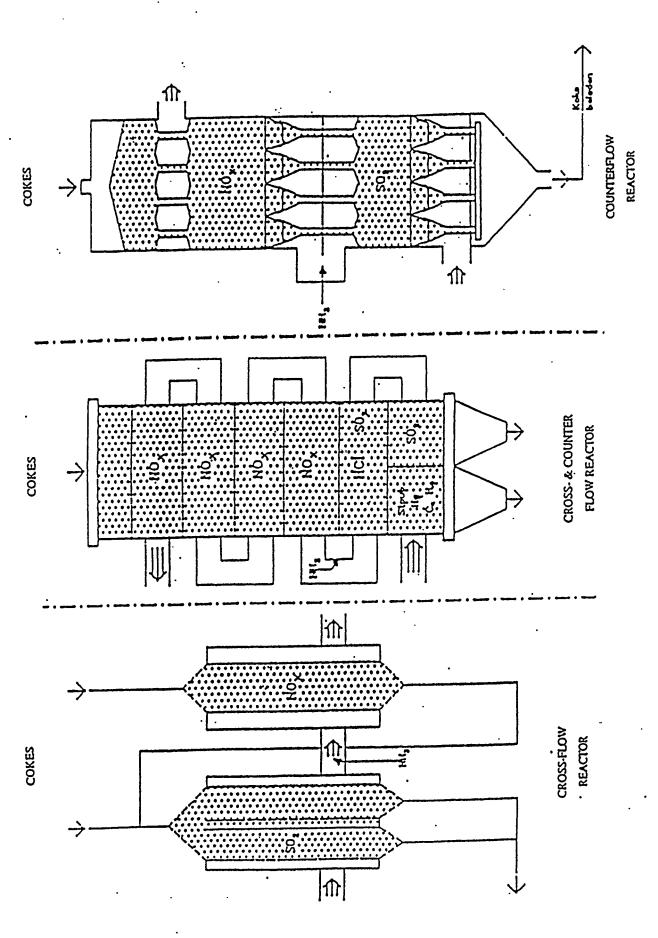


Figure 1. Various moving bed filter configurations.

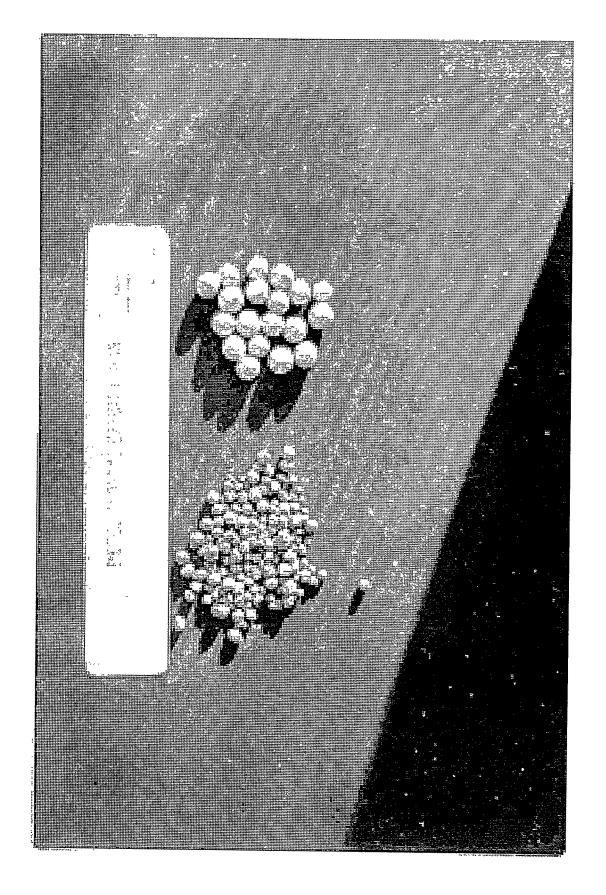


Figure 2. Photograph of Sorbalit Pellets

Scrap tire pyrolysis: experiment and modelling

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INTRODUCTION

Pyrolysis of waste, usually organic solids like tires, plastics or composite materials, is an alternative thermal waste treatment technology. Three main physical and chemical mecanisms -i.e.: chemical kinetics, internal heat transfer and external heat transfer - have to be considered when modelling the degradation of solid waste particles. Because of the lack of physical properties for wastes most of the models described in the literature use basic data obtained on the pyrolysis of coal, wood and biomass. In this work, we report basic information on the thermal degradation of tire samples at small scale: Thermogravimetric analyser (TGA) and differential scanning calorimeter (DSC), as well as direct and indirect measurements of thermal and physical properties (thermal conductivity of the tire and of the char, porosity, density, specific heat). Pyrolysis experiments on tire samples are performed in an imaging furnace. The experimental results are compared to theoretical values deduced from models that take into account physical property measurements.

ANALYSIS

Works on the pyrolysis of coal, wood and biomass have led to numerous model descriptions taking into account heat and mass transfer in large solide particles undergoing pyrolysis reactions. Pyle and Zaror¹ studied two major dimensionless groups: the Biot number (Bi) which compares external convection and internal conduction and the Pyrolysis number (Py), which compares internal heat conduction transfer and chemical kinetics. Internal convection is assumed negligible and external heat transfer (convection and radiation) may be described by a third-order boundary condition. Based on this analysis, simplified models can be developed for design or modelling of reactors. A fundamental study of wood pyrolysis by Kansa and co-authors² takes into account heat conduction, internal convection, mass, energy and momentum equations for the solid and the released gas. The influence of the Peclet number (Pe), which compares internal conduction and internal convection, is studied. In most pyrolysis models, internal convection is neglected.

Chemical kinetics

The pyrolysis reaction in a solid (wood, biomass particle, coal, etc.) is described by many authors^{1,3-5} as a source term in the local energy equation:

$$p = -\Delta H \frac{\partial \rho_s}{\partial t} \tag{1}$$

where

 $p = source term in W.m^{-3}$

 ΔH = overal heat of reaction during the pyrolysis process in J.kg⁻¹

 $\frac{\partial \rho_s}{\partial t}$ = local mass loss in kg.m⁻³.s⁻¹

The local mass loss is determined from the overall mass loss obtained by TGA. Several methods are described to determine the kinetic law from non-isothermal thermogravimetric analyser (TGA) measurements⁶. Other authors¹⁻² assume a simple Arrhenius decomposition equation:

$$\frac{\partial \rho_{s}}{\partial t} = -Ae^{\frac{-E}{RT}}(\rho_{s} - \rho_{s\infty})^{n}$$
(2)

where $\begin{array}{lll} A & = & \text{frequency factor in s}^{-1} \\ E & = & \text{activation energy in J.mol}^{-1} \\ R & = & \text{gas constant in J.mol}^{-1}.K^{-1} \\ T & = & \text{temperature in K} \\ \rho_s & = & \text{density of the solid at a time t in kg.m}^{-3} \\ \rho_{s\infty} & = & \text{density of the solid at the end of the pyrolysis reaction in kg.m}^{-3} \\ n & = & \text{order of the reaction} \end{array}$

In the case of waste, which are multi-component and heterogeneous materials, little information is available on the chemical reactions involved in pyrolysis. The basic TGA and DSC measurement methods provide a global information about heat and mass balance on a small sample of the material. Some of the reactions may be exothermic, others endothermic, according to the temperature range. Yang and co-authors⁷ studied the degradation of the elastomers which are the main degradable components of waste tires: styrene-butadiene rubber (SBR), polybutadiene rubber (BR) and natural rubber (NR). The mass loss is the algebric sum of the mass losses of the elastomers:

$$\frac{\partial \rho_{s}}{\partial t} = \sum_{i} \frac{\partial \rho_{i}}{\partial t} \tag{3}$$

where

 $\frac{\partial \rho_i}{\partial t}$ = local mass loss of component i in kg.m⁻³.s⁻¹

A first order kinetic equation is used to describe the reaction kinetics:

$$\frac{\partial \rho_{i}}{\partial t} = -A_{i} e^{\frac{-E_{i}}{RT}} \rho_{i} = K_{i} \rho_{i}$$
(4)

where

A_i = frequency factor of the component i in s⁻¹

E_i = activation energy of the component i in J.mol⁻¹

K_i = rate constant of the component i in s⁻¹

density of the component i et a time t in la me⁻²

 ρ_i = density of the component i at a time t in kg.m⁻³

Kim and co-authors calculate K_i and $\frac{\partial \rho_s}{\partial t}$ by fitting the TGA data with Equation (3). The method

requires no preliminary investigation about the initial material composition. It is assumed that tire rubber is constituted of various compounds which undergo independant decomposition reaction in characteristic individual temperature regions. Each decomposition is governed by a first order irreversible independant reaction and the kinetic parameters were obtained by differential methods⁹⁻¹⁰

Even with a realistic description of the mass loss term as a function of temperature, Equation (1) does not take into account the thermal effects at different temperatures.

For waste tire pyrolysis, degradation exhibits an exothermic reaction between two endothermic stages¹¹. Mass loss occurs at 200°C, but thermal degradation (softening and liquefaction) starts almost at ambient temperature. It is thus important to have a better description of the source term used in the energy equation.

Yang and co-authors¹¹ make a clear distinction between the source term induced by thermal degradation (no mass loss) and the source term induced by volatilization of the elastomers. The heat release process is described by a simulative curve of the DTA and DSC enthalpy measurements.

The decomposition source term without mass loss is expressed as:

$$p_{d} = \sum h_{di} \rho_{i} \frac{d\alpha_{i}}{dt}$$
 (5)

where

h_{di} = heat of reaction of component i in J.kg⁻¹

 $\frac{d\alpha_i}{dt}$ = rate of decomposition of component i in s⁻¹

 ρ_i = density of component i in kg.m⁻³

The evaporation source term is expressed as:

$$p_{v} = \sum h_{gi} \frac{d\rho_{i}}{dt}$$
 (6)

where

 h_{gi} = latent heat of component i in J.kg⁻¹

 $\frac{d\rho_i}{dt} = \text{rate of weight loss of component i kg.m}^{-3}s^{-1}$

Heat transfer

When the Pyrolysis number is small, the pyrolysis process is controlled by internal or external heat transfer. The heat transfer problem is often treated as a moving boundary problem ¹³. Among the models which were developed for wood and biomass pyrolysis, integral balance techniques provide approximate analytical solutions ¹⁴. For internal controlled processes (small Biot number), pyrolysis rate is very sensitive to thermal conductivity and a good prediction of the pyrolysis time requires an accurate knowledge of the conductivity of the material and of the char. In the case of waste or heterogeneous materials, little information on the thermal conductivity is available. For the char, the conductivity is dependant on the structural properties of the material itself. The wood-cell arrangement generates a pseudo-periodic porous char. For tire particles, evaporation occurs in two-phase porous medium made of liquid elastomers and solid carbon black. This 'bubbling' mechanism ¹⁵ generates microporosity in the char.

The conduction term in the energy equation can be written:

$$q = -\nabla \cdot (k^* \nabla T) \tag{7}$$

where

 k^* = apparent thermal conductivity in W.m⁻¹.K⁻¹

∇ = gradient (vector) differential operator equivalent to a partial derivative in a one dimensional system

A review of thermal conductivity models in porous media was made by Azizi¹⁶. The apparent thermal conductivity is influenced by the conductivity of the two phases and the proportion of the phases (porosity). But the influence of geometry is important and more information about the microscopic arrangement of the carbon black in the porous structure is needed for a correct description of conductive heat flow. In the litterature about pyrolysis models, the parallel model in which the thermal conductivity

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is assumed to be linearly dependent of the various phases conductivity, is often used without justification¹¹. For large temperature gradients, internal radiative transfer must be considered.

EXPERIMENTS

Kinetics

The experiments were performed with a Calvet-type heat flow Differential Scanning Calorimeter (Setaram DSC 111) coupled with a thermobalance (Setaram ATG 92). The reference material was a fixed amount of a-alumina. The powder tire samples were obtained by cryogenic grinding device (SPEX 6700 Freezer/Mill) giving a particle size range of 100-250 µm small enough to ensure internal isothermal conditions during experiments. A flow of purified argon in both the reference and the sample tube was used to maintain an inert atmosphere and to remove the volatile products. The tire samples were studied from 30°C to 800 °C at a heating rate of 5°C.min⁻¹. The final temperature was maintained for one hour to obtain a complete pyrolysed solid (char). Two experiments were realized in the same conditions with the char and the empty crucible respectively in order to record the char and blank signals. Mass loss curves and mass loss rate curves were continuously measured in each experiment.

Thermal diffusivity measurements

The flash method¹⁷⁻¹⁸ was used to measure the thermal diffusivity of tire and char. A brief thermal impulsion is absorbed on the upper side of the sample and the consequent temperature increasing on the back side is recorded. The sample is maintained in a tubular furnace and measurements can be obtained at various temperature from ambient temperature to 300°C. The measurement requires a disc sample of 30 mm diameter, 4 mm thickness with the two faces parallel (Figure 1).

Imaging furnace

An imaging furnace described elsewhere ¹⁹ (Figure 2) was used as the experimental device to study the internal thermophysical tire properties. A Xenon arc lamp (4,5 kW) is the radiative source of energy. Spheric and elliptic mirrors concentrate the flux at the focus of the furnace. The sample (12 mm diameter, 3 mm thickness pellet) lies on the top of a sliding support allowing to lead it to the focus where its upper side is exposed to the high radiative flux while its lower side is insulated. The carrier gas (N₂, 200 mL.min⁻¹) is swept under the quartz cover (60 mm diameter, 60 mm high) to maintain neutral atmosphere (Figure 3). The time of treatment is easily controlled by the mean of the sliding support. The high energy supplied by the imaging furnace allows a great gradient of temperature to establish inside the solid; otherwise, as a high temperature is quickly reached in the solid, we can assume that the pyrolysis kinetic is not a limitative factor. So, the boundary conditions are easily controlled in this furnace and the reaction rate may be neglected regarding to the internal heat transfer. This experimental device allows to study the internal heat transfer in a solid chip submitted to a pyrolysis reaction.

MODEL

The mass and energy balance equations for a material undergoing pyrolysis reactions may be written:

mass balance

$$\frac{\partial \rho_{\rm s}}{\partial t} + \nabla \cdot (\rho_{\rm g} \, \mathbf{v}) = 0 \tag{8}$$

energy balance

$$\frac{\partial}{\partial t} \left[\rho_{g} u_{g} + \rho_{s} u_{s} \right] + \nabla \cdot (\rho_{g} u_{g} \mathbf{v}) = \nabla \cdot (\mathbf{k}^{*} \nabla T) + \frac{-\partial \rho_{s}}{\partial t} \cdot (\mathbf{h}_{g} - \mathbf{h}_{s})$$
(9)

where

apparent density of gas in kg.m⁻³ ρ_{g}

apparent density of solid in kg.m⁻³

 $\rho_s = u_s, h_s = u_g, h_g = u_g$ internal energy, enthalpy of solid in J.kg⁻¹

internal energy, enthalpy of gas in J.kg-1

Darcy velocity of gas in m.s⁻¹

thermal conductivity of the solid in W.m⁻¹.K⁻¹

Assuming convection negligible ($\mathbf{v} = 0$), Equation (9) becomes:

$$\frac{\partial}{\partial t}(\rho_g u_g + \rho_s u_s) = \nabla \cdot (k^* \nabla T) + (-\frac{\partial \rho_s}{\partial t})(h_g - h_s)$$
(10)

Assuming $\rho_g u_g \ll \rho_s u_s$, we obtain :

$$\frac{\partial(\rho_{s}u_{s})}{\partial t} = \nabla \cdot (k^{*}\nabla T) + \frac{-\partial\rho_{s}}{\partial t} \cdot (h_{g} - h_{s})$$
(11)

The internal energy u_s is the sum of the latent heat L_s and the sensible heat c_sT :

$$\frac{\partial(\rho_s c_s T)}{\partial t} = \nabla \cdot (k^* \nabla T) - L_s \rho_s \frac{\partial \alpha}{\partial t} + \frac{-\partial \rho_s}{\partial t} \cdot (h_g - h_s)$$
(12)

where

 $\frac{\partial \alpha}{\partial t}$ = rate of solid-liquid transformation of the solid (0 < \alpha < 1) in s⁻¹

The Pyrolysis number can be defined by:

$$P_{y} = \frac{k^{*}}{K\rho_{s}c_{s}l^{2}} \tag{13}$$

where

the lowest constant rate for the pyrolysis reactions in s⁻¹ K

dimension of sample in m

Assuming $P_y \ll 1$, the controlling factor is heat transfer and thus the quantities α , $h_g - h_s$ and $\frac{d\rho_s}{2}$ in

Equation (12) are only dependent on temperature T.

To solve Equation (12), an integral balance method can be used (in the 1 dimensional case)

$$\int_{D} \frac{\partial \rho_{s} c_{s} T}{\partial t} dx = \int_{D} \nabla . (k^{*} \nabla T) dx + \int_{D} -\frac{\partial \rho_{s}}{\partial t} (h_{g} - h_{s}) dx + \int_{D} -\frac{\partial \alpha}{\partial t} L_{s} \rho_{s} dx$$
(14)

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where

D = domain of integration (thickness of material which changes over time)

A simplified expression of $\int_D -\frac{\partial \rho_s}{\partial t} (h_g - h_s) dx + \int_D -\frac{\partial \alpha}{\partial t} L_s \rho_s dx$ is given by Kanury and Holve¹⁴ assuming a constant pyrolysis temperature T_c with constant bulk enthalpy change ΔH . Equation (14) reduces to:

$$\frac{\partial}{\partial t} \int_{D} \rho_{s} c_{s} T dx = \int_{D} \nabla . (k^{*} \nabla T) dx + (\rho_{c} - \rho_{0}) \Delta H \frac{dx_{c}}{dt}$$
(15)

where x_c , ρ_c , ρ_0 are defined in Table 1

The boundary conditions are:

- constant radiative heat flux at x=0
- no heat loss at x=1

The solution of this integral equation is given by Kanury and Holve¹⁴ with the following assumptions:

- The thermophysical properties are independent of temperature but depending on density. The properties of non reacted solid are noted with subscript 0 whereas the properties of char are noted with subscript c.
- A linear temperature profil is chosen to integrate the energy balance along the x axis.

The model describes the pyrolysis reactions as a three main phase process: a preheated phase (Figure 4a) corresponds to the increase of the temperature from T_i up to T_c into the depth x_t ; an early-charring (Figure 4b) phase where a continuously thickening char layer x_c exists near the exposed surface and a thermal wave x_t propagates into the solid; and a late-charring (Figure 4c) phase where the char front x_c propagates across the solid to the insulated face.

The basic equations of the model are reported in Table 1.

RESULTS AND DISCUSSION

Kinetics

Typical TGA, DTG (derivative thermogravimetry) and DSC results are illustred on Figure 5 and Figure 6 respectively.

The major composite materials of tire rubber are carbon black, elastomers and processing oils^{7,12}. Three stages of degradation are easily recognized on the DTG curve. Brazier and al.²⁰ attributed the first stage of degradation (150-310 °C) to the evaporation of oil/plastizer included in the tire. The second and third stages correspond to the degradation of elastomers. The temperature of degradation of natural or polyisoprene rubber is about 373 °C while styrene-butadiene copolymers rubber decompose on the 430-450 °C temperature range depending on styrene content. The total weight loss is more than 60 % of the initial mass and the remaining char is constituted of inert carbon black and ash. The thermal degradation takes place between 280-480 °C and is achieved when the temperature reached 500 °C. A pyrolysis temperature T_c of 380 °C was choosen for the Kanury model¹⁴.

The DSC curve on Figure 6 shows an endothermic process at the beginning of the treatment, an exothermic transition between 300-400 °C and finally an endothermic process up to 500 °C. The endothermic energy may be due to the rupture of chemical bonds in the polymer leading to a viscous

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aspect. Sircar and al. 12 attribute the exothermic transition to bond-forming reactions like crosslinking, cyclization. The ΔH reaction energy is calculated in the total range of the study and the value of 713 kJ.kg⁻¹ is used in the model. The specific heat of tire is measured in the degradation region 30°C-200°C and a mean value of $c_0 = 1417 \text{ J.kg}^{-1}$.K⁻¹ is chosen. The specific heat of char is determined in the high temperature region 380°C-800°C where the solid is completely degraded. The value of $c_c = 1779 \text{ J.kg}^{-1}$.K⁻¹ is retained for the model.

Thermal diffusivity measurements

Measurements were made with the flash method both for the tire sample and the char. Values are reported in Table 2

To obtain a good measurement, the shape of the sample should be perfectly cylindrical with plane and parallel faces. But during the pyrolysis process, warping and splitting of the sample may be observed especially at high temperature and high heat flux. An important volume change may in some case be observed (up to twice the initial volume). To obtain correct shaped samples, a tubular electrical furnace was used with mild external conditions. The values of thermal diffusivity were given for ambient temperature. Effets of high temperature and radiation were not studied in this experiment.

Imaging furnace

Experimental results are shown on Figure 7. The overall mass loss of a tire sample is plotted versus time when undergoing a high constant heat flux (100 W.cm⁻²) on one face, the other face being insulated. With this very severe external conditions, temperature rise in the sample is very fast and the main limiting phenomen on is internal heat transfer. Comparison between the model and experience points out the influence of thermal conductivity of the solid char on the duration of pyrolysis. The values taken in the model and reported from the experiments described above are recorded in Table 2. The experimental data may be fitted by a theoretical value k_c = 0.085 W.m⁻¹.K⁻¹, noticeably different from the measured value k_c = 0.14 W.m⁻¹.K⁻¹. Several explanations may be given for this discrepancy : the conditions of pyrolysis were different between the imaging furnace and the electrical tubular furnace which was used to prepare the samples. As a matter of fact, high thermal rates may cause greater volume change ("pop corn effects") and increase porosity inside the char thus lowering the apparent thermal conductivity. The existence of cracks and splits may also have identical effects on the char thermal conductivity.

CONCLUSION

Little data on the thermophysical properties of waste is available. This paper points out the importance of these data in the energy equation: heat of reaction, heat of vaporization, overall enthalpy, reaction kinetics, specific heat, thermal conductivity, temperature range of pyrolysis.

A preliminary set of measurement techniques was presented in the present paper with a special interest on scrap tire pyrolysis. The following results were obtained:

1. Combined DSC and TGA is a powerful technique to take into account mass and energy balances in the pyrolysis reactions. Mass losses, but also overall reaction enthalpy, specific heat of waste and char may be obtained. Thermal effects, endothermic and exothermic, may be observed and put in relation with mass losses.

2. For large tire particles, heat conduction is a dominant effect on the overall behaviour of the pyrolysis reaction, but the determination of thermal diffusivity is not straightforward. According to the experiments described the order of magnitude of the thermal conductivity is 0.085 - 0.14 W.m⁻¹.K⁻¹ depending on the pyrolysis process. These very low values show that char is a very insulating material.

The results presented show the importance of a thorough characterization of solid wastes with respect to pyrolysis processes. Heat conductivity, specific heat and heat of reactions have a strong influence on the overall energy balance as well as the characteristic times of the process. A complete model including competition between internal and external heat transfer effects, reactions kinetics, temperature range effects, based on accurate determination of thermophysical parameters is currently developed and will be presented soon.

- 1. D.L. Pyle and C.A. Zaror, "Heat transfer and kinetics in the low temperature pyrolysis of solids," Chem. Eng. Sci., 39(1): 147 (1984)
- 2. E.J. Kansa, H.E. Perlee and R.F. Chaiken, "Mathematical model of wood pyrolysis including internal forced convection," <u>Combust. Flame</u>, 29: 311 (1977)
- 3 H.C. Kung, "A mathematical model of wood pyrolysis," Combust. Flame, 18: 185 (1972)
- 4 Z. Pan, H.Y. Feng and J.M. Smith "Rates of pyrolysis of Colorado oil shale," <u>A.I.CH.E. J.</u>, 31: 721 (1985)
- 5 C.A. Koufopanos, N. Papayannakos, G. Maschio and A. Lucchesi "Modelling of the pyrolysis of biomass particles. Studies on kinetics, thermal and heat transfer effects," <u>Can. J. Chem. Eng.</u>, 69: 907 (1991)
- 6 W.W. Wendlandt, Thermal analysis, 3rd ed., John Wiley & Sons, New York, 1986, pp 57-82.
- 7 J. Yang, S. Kaliaguine and C. Roy "Improved quantitative determination of elastomers in tire rubber by kinetic simulation of DTG curves," <u>Rubber Chem. Technol.</u>, 66(2): 213 (1993)
- 8 S. Kim, J.K. Park and H.D. Chun "Pyrolysis kinetics of scrap tire rubbers. I: using DTG and TGA," <u>J. Environ. Eng.</u>, july: 507 (1995)
- 9 J.H. Flynn and L.A. Wall "General treatment of the thermogravimetry of polymers," <u>J. Res. Nat. Bur. Standard</u>, 70A(6): 487 (1966)
- 10 P. Kofstad "Oxidation of metals: determinatuion of activation energies," Anal. Chem., 179(Jun. 29): 1362 (1957)
- 11 J. Yang, P.A. Tanguy and C. Roy "Heat transfer, mass transfer and kinetics study of the vacuum pyrolysis of a large used tire particle," Chem. Eng. Sci., 50(12): 1909 (1995)
- 12 A.K. Sircar and T.G. Lamond "Identification of elastomers by thermal analysis," <u>Rubber Chem.</u> <u>Technol.</u>, 345: 329 (1972)
- 13 R.E. Desrosiers and R.J. Lin "A moving-boundary model of biomass pyrolysis," <u>Sol. Energy</u>, 33(2): 187 (1984)
- 14 A.M. Kanury and D.J. Holve "Transient conduction with pyrolysis (Approximate solutions for charring of wood slabs)," <u>J. Heat Trans.</u>, 104: 338 (1982)
- 15 A. Attar "Bubble nucleation in viscous material due to gas formation by a chemical reaction: application to coal pyrolysis," A.I.Ch.E. J., 24(1): 106 (1978)
- 16 S. Azizi: "Conductivité thermique des matériaux poreux humides: mesure par méthode flash et interprétation", Thesis INPL, 25 Mars 1988, Nancy, France.
- 17 A. Degiovanni "Diffusivité et méthode flash," Rev. Gén. Ther., 185: 420 (1977)

- 18 W.J. Parker, R.J. Jenkins, G.P. Buttler, G.L. Abbott "Flash method of determining thermal diffusivity heat capacity and thermal conductivity," <u>J. Appl. Phys.</u>, 32(9) (1961)
- 19 S. Bennini, S. Castillo, J.P. Traverse "Effects of an intense thermal flux on a lignocellulosic material," J. A. A. P., 21: 305 (1991)
- 20 D.W. Brazier and G.H. Nickel "Thermoanalytical methods in vulcanizate analysis II. Derivative thermogravimetric analysis," <u>Rubber Chem. Technol.</u>, 48: 661 (1975)

Table 1. Kanury and al. model

Comments	T_i initial temperature in K T_c pyrolysis temperature in K T_c abscisse in m 1 depth of solid in m $\alpha_0 = \frac{k_0}{\rho_0 c_0}$ thermal diffusivity of tire in m ² .s ⁻¹ time in s t_0 thermal conductivity of tire	in W.m ⁻¹ .K ⁻¹ k _c thermal conductivity of char in W.m ⁻¹ .K ⁻¹	AH enthalpy of pyrolysis reaction in J.kg ⁻¹ ." heat flux on the upper side of solid in W.m ⁻²	ρ _c density of tire in kg.m ⁻³ ρ ₀ density of tire in kg.m ⁻³ $x_c \text{ abscisse of the char front}$ in m
Mathematical model	$\theta \equiv \frac{T - T_i}{T_c - T_i} \xi \equiv \frac{\kappa}{l} \tau \equiv \frac{\alpha_0 t}{l^2} \sigma \equiv \frac{\rho_c c_c}{\rho_0 c_0}$ $= \frac{1}{q} \equiv \frac{\alpha_0 t}{\rho_0 c_0} \chi \equiv \frac{\Delta H(\rho_0 - \rho_c)}{\rho_0 c_0(T_c - T_i)} \kappa \equiv \frac{k_c}{k_0}$ $\xi_T(\tau) = \sqrt{2}\tau$	$\xi_{c}(\tau) = 0$	$\xi_{c}(\tau) = \frac{\kappa \lambda}{\sigma \bar{q}} \left(\sqrt{1 + \frac{2\sigma \bar{q}}{\kappa \lambda^{2}}} \left(\bar{q}\tau - \frac{F(\tau)}{2} \right) - 1 \right)$ with $F(\tau) = \sqrt{4 \left(\tau - \frac{1}{2\bar{q}^{2}} \right) + \frac{1}{\bar{q}^{2}}}$	$\xi_{c}(\tau) = \frac{\kappa \lambda}{\sigma \bar{q}} \left(\sqrt{1 + \frac{2\sigma \bar{q}}{\kappa \lambda^{2}}} \left(\left(\bar{q} - \frac{1}{1 - \xi_{cs}} \right) (\tau - \tau_{s}) + \lambda \xi_{cs} + \frac{\sigma \bar{q}}{2\kappa} \xi_{cs}^{2} \right) - 1 \right)$ with $\xi_{cs} = \xi_{c}(\tau_{s})$
Phase	Adimensional variables	$0 \le \tau \le \tau_0$	Early-Charring phase $\tau_0 \le \tau \le \tau_s$	Late-Charring phase $\tau_s \leq \tau$

Table 2. Property values used in the Kanury and al. model

	Tire	Char	Reference
Conductivity (W.m ⁻¹ .K ⁻¹)	0.35	0.14	Flash method
Specific heat (J.kg ⁻¹ .K ⁻¹)	1417	1779	DSC
density (kg.m ⁻³)	1100	479	Yang ¹¹
Pyrolysis temperature T _c (K)	(553	ATG
Heat of Pyrolysis ΔH (kJ.kg ⁻¹)	713		DSC
Radiative Flux $q_s^{"}$ (W.m ⁻²)	10 ⁶		
Depth of solid 1 (m)	3	10 ⁻³	

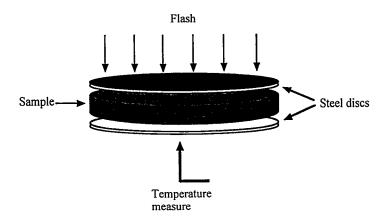


Figure 1. Schematic device of thermal diffusivity measurement by the flash method

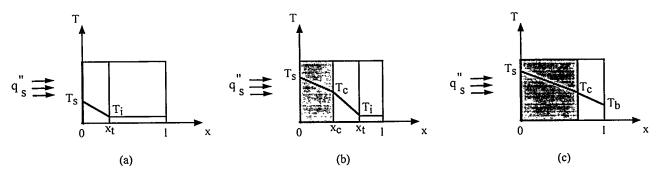


Figure 4. Schematic representation of the Kanury and al. model

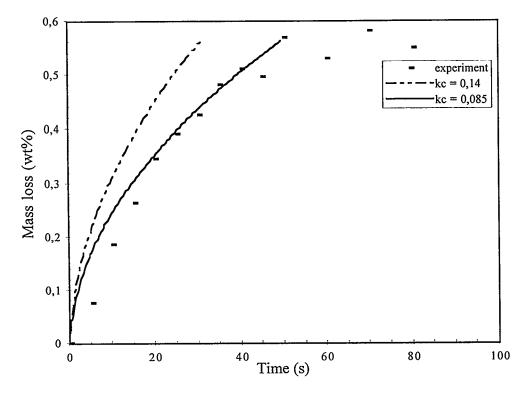


Figure 7. Mass loss vs. time : experimental results (imaging furnace); Kanury and al. model (kc=0.14 and $0.085~W.m^{-1}.K^{-1}$)

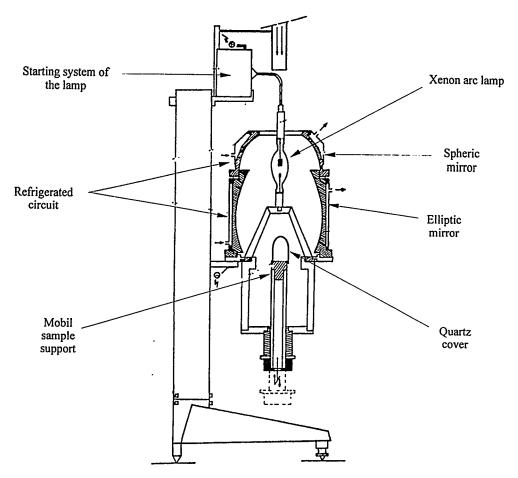


Figure 2. Imaging furnace

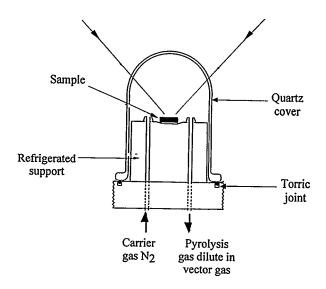


Figure 3. Detailled quartz cover

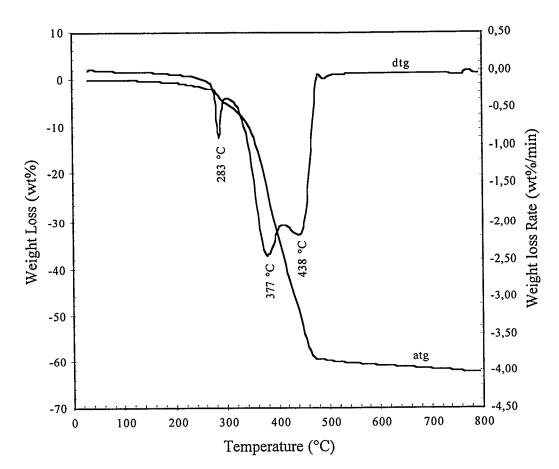


Figure 5. ATG and DTG curves of the pyrolysis of tire

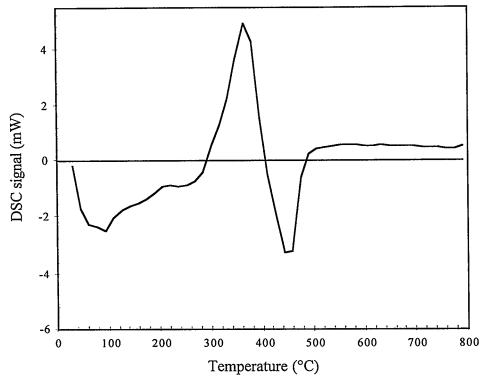


Figure 6. DSC curve of the pyrolysis of tire